CHAPTER IV
MAJOR WATERBODIES OF THE GREAT WATERS:
AN OVERVIEW OF PROGRAMS AND EFFORTS ADDRESSING ATMOSPHERIC DEPOSITION

Section 112(m) of the Clean Air Act specifically designates the Great Lakes, Lake Champlain, Chesapeake Bay, and certain other U.S. coastal waters as waterbodies EPA is to consider in identifying and assessing atmospheric deposition of hazardous air pollutants (HAPs) to the Great Waters. Researchers have found that the Great Waters have been affected by metals, pesticides, toxic chemicals, and nutrients that enter the waters through different pathways, including atmospheric deposition.

This chapter presents information currently known about contamination occurring at the individual Great Waters, including problems or issues that challenge each waterbody, followed by discussion of current strategies or efforts to respond to these concerns. Most of the information has been generated from activities occurring at the Great Lakes and Chesapeake Bay. In the Great Lakes, several initiatives have been introduced in recent years, ranging from research projects to gather quantitative estimates of atmospheric loadings to regulatory and voluntary activities promoting reduction of loadings to the waterbody. Efforts at Chesapeake Bay have focused on developing models to improve characterization of nitrogen and toxic contaminant loadings, as part of major reduction strategies. Research is more limited for Lake Champlain and for the other coastal waters. In these waterbodies, smaller-scale investigations have been carried out to study certain pollutants of concern in their respective waterbodies, and in many cases, comprehensive strategic plans have been developed to address contamination issues.

This chapter is organized by sections on each of these major waterbodies of the Great Waters and divided accordingly:

- Section IV.A presents information available on atmospheric deposition of persistent toxic pollutants into the Great Lakes and the many programs to characterize and reduce loadings;

- Section IV.B describes Lake Champlain and current research to assess atmospheric mercury deposition in the basin;

- Section IV.C discusses the deposition of nitrogen and toxic pollutants to Chesapeake Bay and the related monitoring and modeling efforts; and

- Section IV.D provides an overview of U.S. estuary programs and some major efforts to characterize loadings of nitrogen and toxic pollutants to coastal waters.

Although this chapter describes current data and programs specific to the subject waterbody, much of the information is relevant to other waters as well. For example, those interested in smaller estuaries will gain insight from information presented in the Chesapeake Bay section.

The Great Lakes and Lake Champlain represent two important freshwater systems in the United States. Lakes are sensitive to pollution inputs because they lack any dominant, unidirectional flow, and as a result, there is a slow change of water and a resulting retention of pollutants.

The Great Lakes contain approximately one-fifth of the world's supply of fresh surface water. These lakes have played a vital role in the history and development of the United States and Canada. They are stressed by a wide range of pollution sources associated with the large urban centers located on their shores. Because the Great Lakes system is a relatively closed water system (very large volume, with relatively small water inflows and outflows), many of the pollutants that reach the Great Lakes remain in the system for extended periods of time. For example, Lake Superior replaces all the water in the lake every 191 years, Lake Erie every 2.6 years.

Lake Champlain is located in the northeastern United States, shared by the states of New York and Vermont and the Province of Quebec. Although much smaller in surface area than the Great Lakes, Lake Champlain is still one of the largest freshwater lakes in the United States and its natural resources are important to the local economy. The Lake Champlain basin, or watershed, is much larger relative to its water surface area than the Great Lakes, and so watershed throughput is much more of an issue for Lake Champlain. Toxic pollutants are an issue of wide public concern in the Lake Champlain Basin, due in large part to fish consumption advisories for PCBs and mercury issued by both New York and Vermont, and the potential impact on drinking water and the Lake's many other uses.

"Coastal waters," for the purposes of CAA section 112(m), are defined as those estuaries designated for the National Estuary Program (pursuant to section 320(a)(2)(A) of the federal Water Pollution Control Act) or designated for the National Estuarine Research Reserve System (pursuant to section 315 of the Coastal Zone Management Act). Chesapeake Bay is identified by name in section 112(m).

Estuaries occur where rivers empty into the ocean, mixing together fresh water and salt water, and creating an ecosystem distinct from, and often more productive than, either fresh or salt water systems. Estuarine waters include bays, sounds, marshes, swamps, inlets, and sloughs. These environments are characterized by varying degrees of salinity, high turbidity levels, and complex water movement affected by ocean tides, river currents, and wind. Estuaries are critical coastal habitats that serve as spawning grounds, nurseries, shelters, and food sources for many different species of shellfish, fish, birds, and other wildlife. The leading environmental problems in estuarine systems at present are eutrophication,[10] contamination by toxic chemicals and pathogens (disease-causing organisms), over-harvesting, and loss of habitat.

In 1975, Chesapeake Bay became the nation's first estuary to be targeted for protection and restoration. Over the past decade, other coastal programs, such as the National Estuary Program, the National Estuarine Research Reserve System, and the Gulf of Mexico Program, have been established to protect and restore water quality and living resources in U.S. estuaries and coastal waters. Chesapeake Bay was also among the first estuaries where atmospheric sources of nutrients and toxic pollutants were recognized as significant inputs to the waterbody. Recently, research on other U.S. coastal waters has begun to evaluate the loadings of nutrients and toxic pollutants to their watersheds from atmospheric sources. The Great Waters program has focused primarily on Chesapeake Bay for estuarine issues, and has found that information developed for this waterbody is generally applicable to several other East Coast estuaries, when

[10] As discussed in Section II.D and in this chapter, eutrophication is over-enrichment of waters that is characterized by algae blooms, turbid waters, and low or no dissolved oxygen conditions.

adjustments are made for respective waterbody's physical, chemical, and geomorphological characteristics.

IV.A The Great Lakes

The Great Lakes, comprised of Lakes Superior, Michigan, Huron, Erie, and Ontario, are an important part of the physical, cultural, and industrial heritage of North America (see Figure IV-1). The Great Lakes ecosystem, the interacting components of air, land, water, and living organisms, including humans, is one of the largest surface systems of freshwater on earth. This ecosystem contains 18 percent of the world's freshwater supply and 95 percent of the surface freshwater within the United States. Only the polar ice caps and Lake Baikal in Siberia contain more freshwater than the Great Lakes. By virtue of their size, the Great Lakes affect the climate of the surrounding region. Areas of Michigan, Ontario, and New York generally have warmer, though snowier, winters than other parts of North America at similar latitudes because, as a result of little continual current, the lakes retain a large amount of heat. In spring and early summer, the lakes are slow to warm, thereby keeping the nearby land areas cool.

The Great Lakes sustain a rich diversity of fish, birds, and other wildlife. Native fishes important for commercial and recreational harvest include lake trout, lake whitefish, and walleye. Non-native species such as smelt, white perch, brown trout, rainbow trout, and several Pacific salmon species also contribute substantially to the total annual fish harvest. Approximately three million waterfowl follow the Atlantic and Mississippi flyways through the Great Lakes basin each year. Native animals include deer, fox, moose, wolves, beaver, mink, and muskrat. In addition, the Great Lakes ecosystem supports more than 100 globally endangered or rare species (Nature Conservancy 1994).

Economic Highlights of the Great Lakes

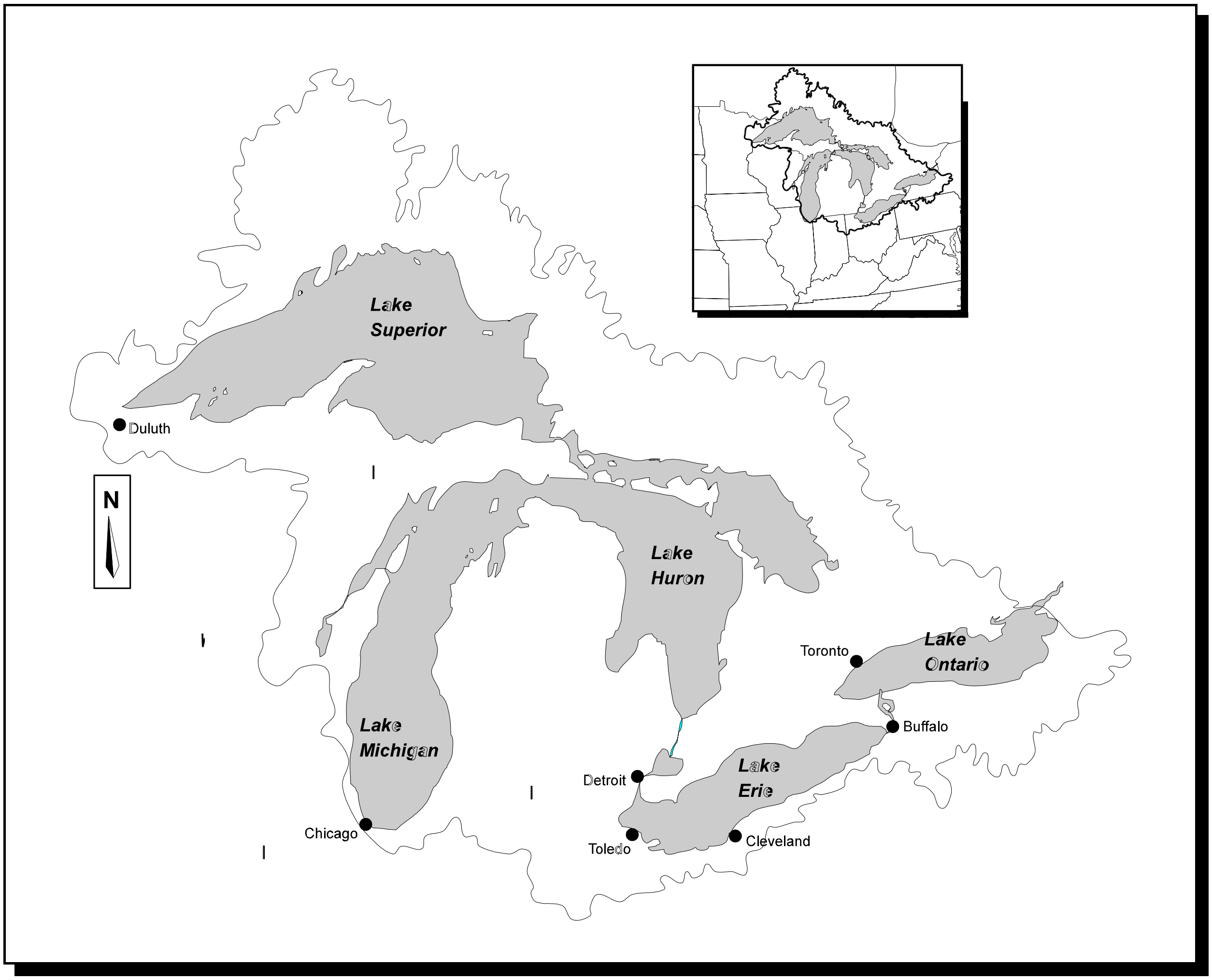
- Approximately 11% of total employment and 15% of manufacturing employment of combined U.S. and Canadian workers are located in the Great Lakes basin.
- Trade between Canada and the eight Great Lakes States in 1992 was valued at $106 billion (56.2% of the U.S.-Canada total).
- An estimated 900,000 to 1 million U.S. and Canadian boats operate each year, resulting in a direct spending impact on the regional economy of more than $2 billion.
- About 2.55 million U.S. anglers fish the Great Lakes; total trip-related and equipment expenditures were $1.33 billion in 1991.

Source: Allardice and Thorp 1995.

The Great Lakes basin is home to more than 33 million people, including 10 percent of the U.S. population and 25 percent of the Canadian population. Over 23 million of these people depend on the Great Lakes for drinking water. Industries use the water to make products, to cool manufacturing processes or power generation equipment, and to ship raw materials and finished products. Residents and visitors alike enjoy an abundance of recreational activities, including boating, swimming, fishing, sightseeing, camping, and hiking.

The concentration of human activities in the Great Lakes basin (e.g., manufacturing, transportation, agriculture, fishing) imposes stresses on the ecosystem and has prompted significant concerns for the health and well-being of the human residents. Many of the major stressors and resultant effects were documented in the First Great Waters Report to Congress. The current report builds on this information and presents some of the potential problems that may affect the Great Lakes basin from the perspective of the CAA, and major programs that are underway to address those problems.

FIGURE IV-1
Great Lakes Basin



Physical Features of the Great Lakes						
	Superior	Michigan	Huron	Erie	Ontario	Totals
Volume (km^3)	12,100	4,920	3,540	484	1,640	22,684
Maximum Depth (meters)	406	282	229	64	244	NA
Water Area (km^2)	82,100	57,800	59,600	25,700	18,960	244,160
Land Drainage Area (km^2)	127,700	118,000	134,100	78,000	64,030	521,830
Retention Time (years)	191	99	22	2.6	6	NA

NA = not applicable

The remainder of Section IV.A presents:

- Current knowledge and recent measurements of atmospheric levels and deposition of toxic pollutants to the Great Lakes;

- Information on major activities/programs currently in progress to assess atmospheric deposition of air pollutants to the Great Lakes;

- Efforts supported by the United States, as well as Canada, to reduce and mitigate atmospheric emissions in the Great Lakes basin; and

- Brief discussion on current information gaps, and future research needs to improve understanding of atmospheric deposition of pollutants into the Great Lakes.

Common Terminology for Pollutant Movement in a Waterbody

FLUX

Transport of a chemical across an interface (e.g., between air and water) for a given area and time, accounting for both inputs and outputs. Net flux is equal to all positive loadings minus all negative loadings.

INPUTS (positive loading)

Wet Deposition: Gases and particles carried in precipitation (rain, snow, sleet) and deposited on land and water surfaces.
Dry Particle Deposition: Pollutants, bound to particles, deposited on land and water surfaces in the absence of precipitation.
Gas Absorption: Gaseous form of pollutants crossing air-water interface into the water (portrayed as a positive number in a flux calculation).
Waterborne Discharge: Pollutants discharged directly to water (e.g., by industrial discharge, urban storm-runoff).
Tributary Loading: Pollutants entering waterbody through connecting channels, streams, and rivers.

OUTPUTS (negative loading)

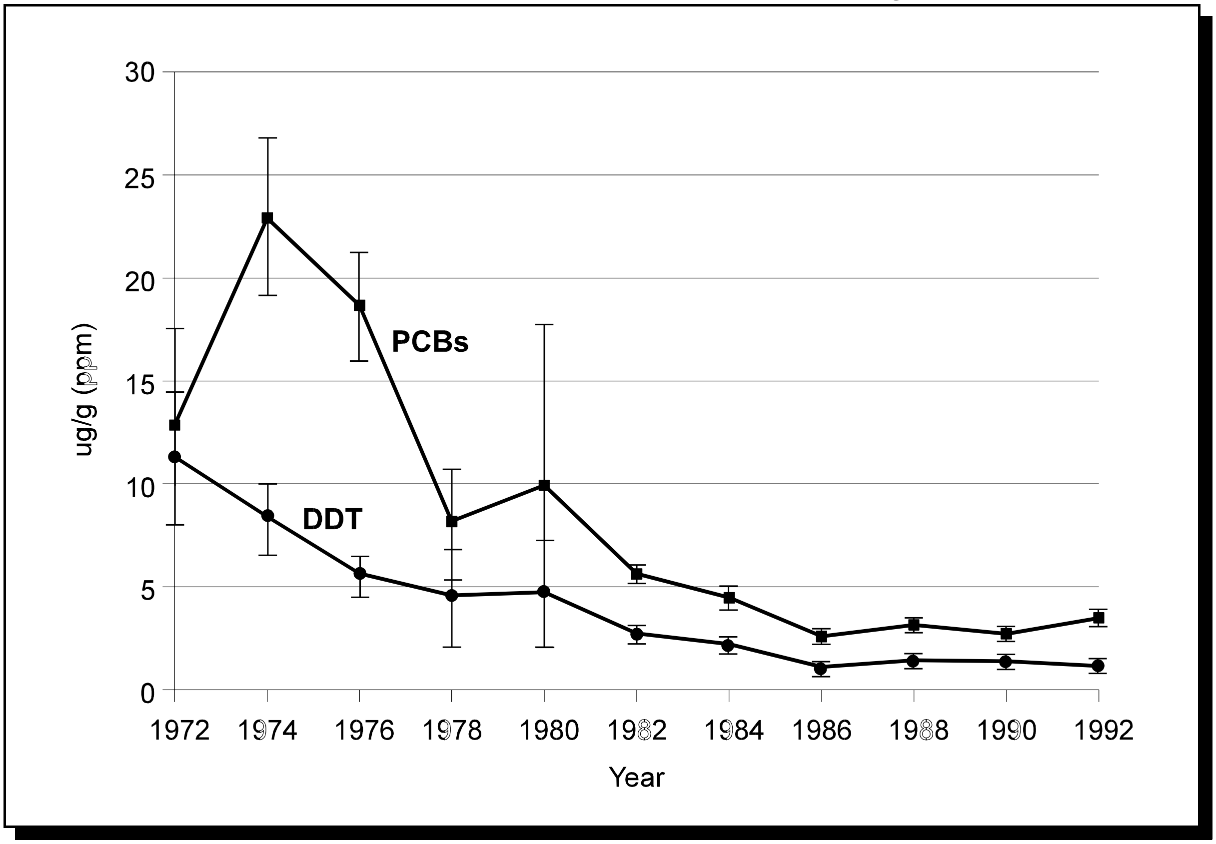
Volatilization or Gas Evasion: Gaseous form of pollutants crossing air-water interface into the air (portrayed as a negative number in a flux calculation).
Sedimentation: Settling of particles by gravity to bottom sediments.
Outflow: Pollutants flowing with water out to rivers or to the ocean.

Atmospheric Deposition of Great Lakes Contaminants

Hundreds of anthropogenic chemicals have been identified in the Great Lakes ecosystem. High levels of certain bioaccumulative pollutants remain in certain fish and wildlife species, and fish advisories have been issued by many Great Lakes states for several pollutants of concern, specifically chlordane, dioxins, mercury, PCBs, and toxaphene (specific advisories are listed in Appendix B). For example, although concentrations of PCBs and DDT in Lake Michigan lake trout are currently about one-tenth of those of 20 years ago (Figure IV-2), the concentrations are still at levels that warrant issuance of public health advisories regarding the consumption of these fish. Advisories may especially apply to specific subpopulations, such as children and women who are pregnant or anticipate bearing children.

The pollutants of concern have been associated with health problems in certain fish and wildlife species, although with the decline of some pollutant levels, many species may be recovering. For example, the number of double-crested cormorants living on the Great Lakes has increased more than 20-fold during the past 15 years. Prior to this, numbers of these fish-eating birds declined during the 1970s due to reproductive failure from DDE-induced egg shell thinning. Health problems persist for fish and wildlife in certain locations, particularly in waters

Figure IV-2
PCBs and DDT in Lake Trout from Lake Michigan



with highly contaminated bottom sediments, and for predators high in the food web, such as lake trout, mink, and bald eagles.

During the 1980s, studies in the Great Lakes showed that atmospheric deposition may be a major route of introduction of a number of pollutants to the Great Lakes. For example, atmospheric transport of toxaphene was implicated when the insecticide was found in fish in Lake Siskiwit, located on an island in Lake Superior Because the elevation of Lake Siskiwit is above that of Lake Superior, it does not receive any groundwater from Lake Superior and thus pollutant input. This pesticide was used mainly on cotton crops in the southern United States, prior to its cancellation in 1982 (McVeety and Hites 1988).

As a result of this and other findings, the United States and Canada established a joint monitoring network called the Integrated Atmospheric Deposition Network (IADN). The IADN is designed to assess the magnitude and trends of atmospheric deposition of target chemicals to the Great Lakes, and to determine emission sources wherever possible. The program responds to the Great Lakes Water Quality Agreement (GLWQA) between the United States and Canada, specifically to the needs of Annex 15 which addresses issues concerning airborne contaminants in the Great Lakes basin. A more detailed discussion of the rationale, design, and results of the IADN, as well as the uncertainties that exist in calculating atmospheric deposition estimates, is presented in the following subsection (Program Actions to Characterize Atmospheric Contamination in Great Lakes).

The first consensus data report for IADN (Eisenreich and Strachan 1992) revised and improved the very broad estimates of atmospheric deposition of toxic contaminants that were previously compiled (Strachan and Eisenreich 1988). More recent data were incorporated into deposition estimates for 1994 and compared to the earlier results (Hoff et al. 1996), as shown in Table IV-1. A general decline in some pollutant levels is suggested from the estimates in Table IV-1. Average estimated atmospheric loadings of certain pollutants to the five Great Lakes between 1991 and 1993 are presented in Table IV-2. The data in Tables IV-1 and IV-2 may not be comparable because the estimates represent measurements at different time frames. It is important to recognize that the values presented in these tables are based on preliminary data and limited samples. Furthermore, contributions from water inputs and outputs are not included. As such, overall loadings to the lakes cannot be established from these data alone.

TABLE IV-1
Atmospheric Loading Estimates for Selected Pollutants (kg/year) in the Great Lakes

Pollutant of Concern	Superior	Michigan	Huron	Erie	Ontario
PCBs[a] (Wet and Dry)					
1988	550	400	400	180	140
1992	160	110	110	53	42
1994	85	69	180	37	64
PCBs[a] (Net Gas Transfer)[b]					
1988	-1900	-5140	-2560	-1100	-708
1994	-1700	-2700	--	-420	-440
DDT (Wet and Dry)					
1988	90	64	65	33	26
1992	34	25	25	12	10
1994	17	32	37	46	16
DDT (Net Gas Transfer)					
1988	-681	-480	-495	-213	-162
1994	30	67	--	34	13
Benzo(a)pyrene (Wet and Dry)					
1988	69	180	180	81	62
1992[c]	120	84	84	39	31
1994	200	250	--	240	120

-- Not determined or reported.
[a] Data presented for PCB congeners 18, 44, 52, and 101 (each with 3-5 chlorines in chemical structure).
[b] The convention is to assign a negative number to loss of pollutant from the lake (i.e., volatilization). Thus, the resulting number expresses the mass of a pollutant going into or coming out of the lake per year (i.e., a positive net gas transfer indicates a net input of the pollutant to the lake and a negative net gas transfer indicates a net loss or output from the lake).
[c] Data from 1992 may represent an underestimation in the measurement of benzo(a)pyrene.

Sources: Eisenreich and Strachan 1992; Hillery et al. 1996; Hoff et al. 1996; and Strachan and Eisenreich 1988.

Atmospheric loadings of pollutants are calculated using atmospheric concentration data gathered by IADN and estimates for various parameters such as lake surface area. The wet deposition data are based on estimated annual precipitation rates, and do not use actual rainfall amounts. Although uncertainties exist for the parameters which can lead to some degree of error, the atmospheric deposition estimates are based on the best scientific data currently available.

The remainder of this subsection presents atmospheric concentration and deposition data collected primarily from IADN on some pollutants of concern, as well as information on current trends of pollutant deposition. The pollutants of concern discussed include PAHs, PCBs, pesticides (e.g., DDE, DDT, lindane, toxaphene), and trace metals (e.g., lead, mercury). PCBs,

TABLE IV-2
Average Estimated Atmospheric Loadings[a] of Selected Pollutants to the Great Lakes (kg/year) (1991-1993)

Atmospheric Process[b]	Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
PCBs					
Wet deposition	58	52	180	21	58
Dry deposition	27	16	---	16	5.7
Net gas transfer[c]	-1700	-2700	---	-420	-440
Absorption	320	390	---	340	130
Volatilization	-2000	-3100	---	-760	-560
Dieldrin					
Wet deposition	21	58	10	28	11
Dry deposition	7.4	8	---	5.6	1.7
Net gas transfer	-780	---	---	-610	-320
Absorption	120	200	---	67	43
Volatilization	-910	---	---	-680	-370
DDE					
Wet deposition	2.6	3.8	10	4.6	4.5
Dry deposition	0.4	0.5	---	0.5	0.2
Net gas transfer	---	---	---	---	-170
Absorption	10	26	---	14	12
Volatilization	---	---	---	---	-180
Lindane					
Wet deposition	62	65	140[d]	46	51
Dry deposition	0.6	1.1	---	0.4	0.2
Net gas transfer	140	1200	---	61	16
Absorption	340	1400	---	180	72
Volatilization	-200	-140	---	-110	-56
Benzo(a)pyrene					
Wet deposition	140	170	---	180	56
Dry deposition	58	77	---	63	60
Net gas transfer	87	---	---	---	---
Absorption	100	92	---	51	7.5
Volatilization	-17	---	---	---	---

[a] Values calculated from atmospheric loading equations, and based on atmospheric concentration data collected from Integrated Atmospheric Deposition Network (IADN); summarized from Hoff et al. (1996).
[b] Wet deposition based on estimated annual precipitation rates, and does not use actual rainfall amounts. Dry deposition represents only data for particle form of pollutant (i.e., gaseous form included in absorption values).
[c] Net gas transfer is the sum of gas absorption and volatilization. Water concentration data are taken from past literature and compared with the more recent air measurements, which may lead to some potential error in gas transfer estimates. Values for net gas transfer are rounded off and thus estimates may not add up in the table.
[d] High estimated value may be due to very limited number of samples for 1992 season and should be reconsidered as more data become available (Hoff et al. 1996).
--- Not determined or reported.

toxaphene, and mercury are given greater focus because fish advisories are currently issued for these pollutants. Fish advisories also exist for chlordane and dioxins for the Great Lakes, but they are not addressed in this section because recent information is limited. Potential sources of the pollutants are also discussed, as well as uncertainties in the data.

PAHs

Polycyclic aromatic hydrocarbons (PAHs), a subset of POM, are a class of semi-volatile compounds produced in combustion processes and are widely distributed in the environment. As indicated in the First Report to Congress, approximately 72 to 96 percent of the total annual loading of one common PAH, benzo(a)pyrene, to Lakes Superior, Michigan, and Huron is attributed to atmospheric deposition.

PAHs are detected both in the gaseous and particulate phases, but some of the most toxic PAHs are largely in the particulate phase in the atmosphere. For the most toxic PAHs, dry deposition is generally the main route of deposition to the lakes (Hoff and Brice 1994). For benzo(a)pyrene, however, wet deposition seems to be the major source of atmospheric loadings to Lake Michigan in all seasons of the year. The IADN data suggest that, for Lake Superior, the net movement of the gaseous phase benzo(a)pyrene is largely to the water; data are limited for the other lakes (see Table IV-2). Comparing recent wet and dry deposition values with historic data, the loading of benzo(a)pyrene to the lakes appears to have increased (Table IV-1). However, the 1992 finding may be attributed to an underestimation in the measurement of benzo(a)pyrene (Hoff et al. 1996).

A recent study found that total wet and dry deposition for benzo(a)pyrene was 50 times higher at an urban site (Chicago) than at remote IADN sites in Lakes Michigan and Superior (Sweet and Harlin 1996). The investigators concluded that large areas of Lake Erie, Lake Ontario, and southern Lake Michigan have elevated PAH deposition rates due to emissions from nearby urban areas. Although the total deposition of PAHs are lower in rural than urban sites, the relative amounts of the individual PAHs (i.e., relative ratios of the individual PAHs) is very similar at urban and nonurban sites, suggesting that little chemical degradation occurs during transport of PAHs from urban source areas to rural and remote sites several hundred kilometers away.

PCBs

PCBs are a class of highly toxic, persistent, and bioaccumulative chemical compounds. PCBs in Great Lakes fish have long been linked to developmental and growth problems in infants born to women who regularly consumed PCB-contaminated fish in the late 1970s. PCBs were produced from 1927 to 1977 for the purpose of insulating and cooling electrical equipment. In the late 1970s, Monsanto Company, sole manufacturer of PCBs in the United States, voluntarily stopped production of PCBs. Estimates suggest that 282 million pounds of pure PCBs -- 20 percent of PCBs ever produced -- were still in service at the end of 1988.

PCBs manufactured before production was stopped are still found in the Great Lakes. They are present in older commercial and industrial equipment (e.g., transformers, capacitors). There are no phaseout deadlines that require removal of the equipment to avoid breakage and release, although this equipment is tightly regulated under the Toxic Substances Control Act (TSCA). As a result of past use and disposal practices, PCBs may reside in sediments in surface waters and in other areas, such as waste sites. As the contaminated sediment is disturbed, the

PCBs may be re-released and resuspended in the water, allowing for continued bioaccumulation in Great Lakes fish. Remediation programs are in-place to address PCB-contaminated waste sites. Other continuing PCB sources include unregulated sources that potentially contain PCBs and releases, as well as releases by those PCB owners who are not aware of the presence of PCBs or of the special management requirements for PCB-containing equipment.

Despite the fact that PCBs are one of the most tightly regulated and controlled group of pollutants under federal regulatory programs, fish consumption advisories still exist for PCBs in all five of the Great Lakes (see Appendix B). For example, although PCB levels have declined in Lake Michigan water, there has been a constant or increasing level of PCBs in some Lake Michigan fish in the last few years, possibly due to resuspension from sediment or from changes in the Lake Michigan food chain (see Section II.B).

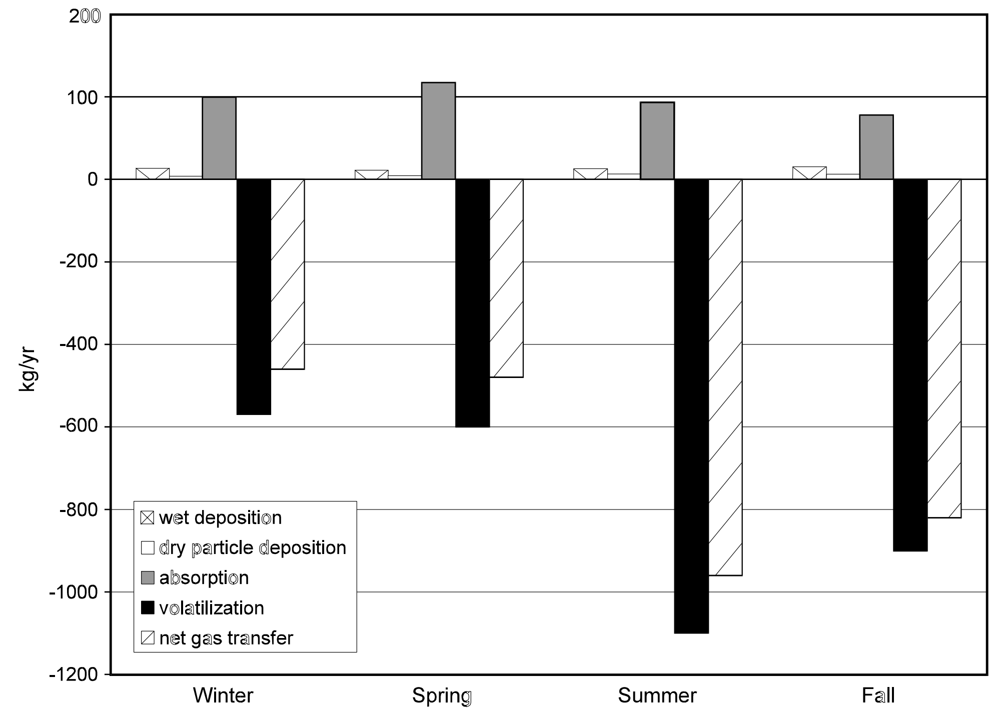
Volatilization is the dominant mechanism in air-water gas exchange of PCBs (Table IV-2). Volatilization of PCBs from the Great Lakes is estimated to be as high as 3,100 kg/year for Lake Michigan and 2,000 kg/year for Lake Superior. In contrast, estimates of wet deposition of PCBs are less than those for volatilization and are nearly the same for Lakes Superior, Michigan, and Ontario (52-58 kg/year) with Lake Erie showing a lower rate (21 kg/year) and Lake Huron showing the highest rate (180 kg/year) (Table IV-2). Dry deposition rates of PCBs are similar in Lakes Superior, Michigan, and Erie (16-27 kg/year), with Lake Ontario showing a lower rate (5.7 kg/year). As presented in Table IV-1, from 1988 to 1994, wet and dry deposition of PCBs to each of the Great Lakes has decreased. Therefore, the net loss to the atmosphere would suggest that the amount of PCBs in water is declining. Tables IV-1 and IV-2, however, represent data only for the atmospheric movement of pollutants and therefore, do not indicate the waterborne inputs to each lake (such as particles in the water, industrial water discharges, and especially urban storm-runoff which goes directly into the lakes). Also, this study on gas exchange is preliminary and there are considerable uncertainties in the estimates. Additional work addressing the uncertainties and other routes of pollutant movement may give a different balance, although the importance of gas exchange is clear.

Wet and dry deposition of PCBs are similar over seasons, while net gas exchange is highly seasonal, exhibiting much greater effect with high temperatures (Figure IV-3). To date, no studies on seasonal variation in PCB concentrations in water have been published to compare with the seasonal atmospheric loading data. As stated earlier, uncertainties exist in calculating deposition estimates, since some estimates are based on rough approximations or assumptions using the best science available at this time.

PESTICIDES

Volatilization of the pesticides dieldrin and DDE (a metabolite of DDT) in many of the Great Lakes is a significant process. Net atmospheric loading is negative, indicating that movement of these two pesticides between air and water is mostly volatilization (Table IV-2). Fluctuations in gas equilibrium conditions may be influenced by the water concentration data, differences in temperature, and/or errors in the Henry's Law constant used in calculating gas movement. For example, DDT net gas transfer estimates are hindered by difficulties in obtaining precise water concentration data because DDT levels in the lakes are close to the analytical detection limit. From Table IV-1, DDT wet and dry deposition loadings declined between 1988 and 1992, but rose slightly for all lakes except Lake Superior in 1994 (Hillery et al. 1996).

FIGURE IV-3
Seasonal Atmospheric Loadings of PCBs in Lake Michigan (1994)



Wet and dry deposition of lindane appeared to be fairly uniform across all lakes (see Table IV-2). Gaseous lindane generally seemed to be in equilibrium within Lakes Erie and Ontario, while gas absorption is the dominant mechanism in air-water exchange for Lakes Superior and Michigan (Table IV-2). The net gas transfer of lindane in Lake Michigan is into the lake in the winter and spring and out of the lake in the summer and fall. For many pesticides, gas transfer is strongly dependent on seasons, with net outputs in the summer and net inputs in the winter (Achman et al. 1992; Hoff et al. 1993; McConnell et al. 1992; Ridal et al. 1996).

Toxaphene in the Great Lakes Basin. Toxaphene, a semi-volatile insecticide containing a mixture of chlorinated bornanes (class of aromatic hydrocarbons), has been recognized as one of the contaminants with the highest concentrations in Great Lakes fish (Ribick et al. 1982; Schmitt et al. 1981, 1985, 1990). Because of its volatility and persistence, toxaphene is still widely distributed through the atmosphere, even though it is no longer used in the United States (Rapaport and Eisenreich 1986). Toxaphene was been found to be a major contaminant in lake trout and whitefish from Siskiwit Lake on Isle Royale, Lake Superior (De Vault et al. 1996) (see sidebar).

Toxaphene in Lake Trout

Since 1991, the state of Michigan has issued a consumption advisory for Siskiwit lake trout from Lake Superior based on exceedance of the FDA's 5.0 ppm action level for toxaphene. In 1995, the Canadian Province of Ontario issued fish consumption advisories for several different species in Lake Superior and upper Lake Huron, triggered by their toxaphene levels and a lowering of Health Canada's action level for toxaphene to 0.2 ppm.

Toxaphene's discovery on pristine Isle Royale, exposed only to atmospheric deposition, seemed indicative of long-range transport via the atmosphere since it had been used primarily as a pesticide in the southern United States (Hoff et al. 1993). This hypothesis has been supported by studies that found toxaphene concentrations in Canadian air masses that had originated in the southern United States. However, there is also evidence to suggest that some of the toxaphene found in Lake Superior and northern Lake Michigan may have local origins. A study that analyzed fish from rivers in the southeastern United States, the Great Lakes, and Isle Royale, collected during 1982, found differences in composition of toxaphene in fish between sites, suggesting that potential local influence may be important, rather than long-distance atmospheric transport from the southeastern United States to the Great Lakes (Petty et al. 1987).

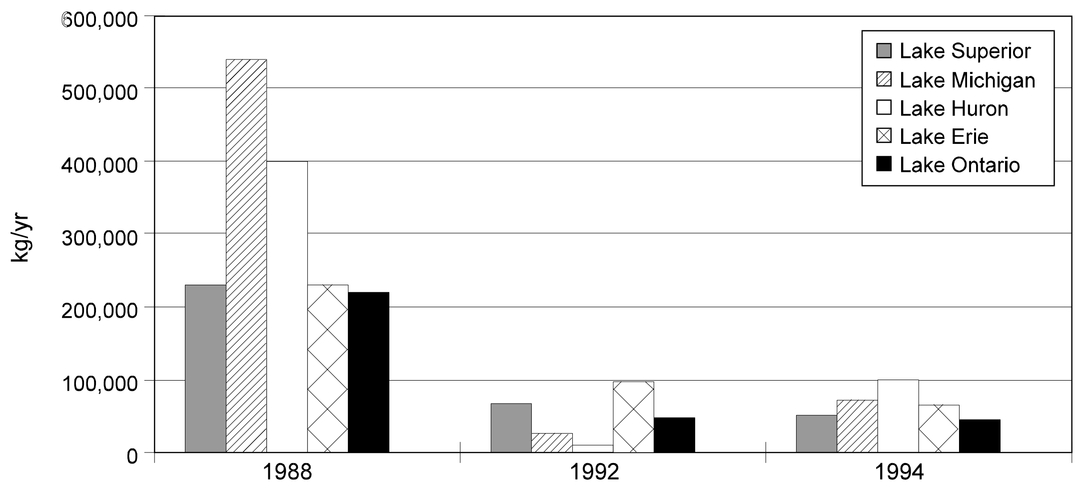
EPA recently supported monitoring of toxaphene in Great Lakes fish and sediment which has revealed two trends. First, there has been a statistically significant decline in the concentration of toxaphene in fish from most waters, as might be expected following reduced use and later cancellation of the pesticide. Second, there has been no discernible decline in toxaphene levels in Lake Superior lake trout; toxaphene levels are higher than levels of other measured contaminants in fish from anywhere in the Great Lakes.

At this time, there are several hypotheses for the relatively elevated levels of toxaphene observed in Lake Superior and northern Lake Michigan. First, the continued use of toxaphene by other countries and subsequent atmospheric transport to the Great Lakes basin may increase levels. Another possibility is the previous local use of the pesticide. Toxaphene was once used to kill undesirable fish communities (Lockhart et al. 1992; Stern et al. 1993). This practice occurred in parts of Canada and the northern United States for fish restocking on small glacial lakes; it was applied to at least 80 lakes during the 1950s and 1960s in Wisconsin (Hughes 1968). It has also been proposed that Lake Superior lake trout may be slower to reflect a decrease in contaminant levels in their food web due to their greater age; however, Glassmeyer et al. (1997) found that toxaphene levels in Lake Superior fish were still elevated compared to levels in fish from the other lakes. Another explanation that has been suggested is that toxaphene persists longer in colder, less productive waters such as Lake Superior. Finally, the high toxaphene levels may be a result of the release of toxaphene into the waters as a byproduct in the production of paper; there are 74 pulp and paper mills that directly discharge to all the Great Lakes (IJC 1995), with the paper industry most concentrated near Lake Superior and upper Lake Michigan (Green Bay).

TRACE METALS

A number of trace metals are of concern in the Great Lakes, though new data are limited. Data from 1994 suggest that wet deposition is the dominant atmospheric transport mechanism for trace metals to the Great Lakes (Hoff and Brice 1994). The most consistent trend in the deposition of trace metals was the reduction in lead in 1994 compared with 1988 values for all the lakes (Figure IV-4). This finding is not surprising given the phaseout of leaded gasoline in the United States beginning in the 1970s and accelerating in the mid-1980s. The gaseous phase of lead is assumed now to be negligible. Arsenic deposition also has decreased. The reason for this finding is not as clear but it has been hypothesized that process changes by Noranda, a major emitter of arsenic in Canada through mining, smelting, and refining of metal products, may have led to the decline.

FIGURE IV-4
Atmospheric Loadings of Lead to the Great Lakes (1988-1994)



Mercury in the Great Lakes Basin. Currently, six of the eight Great Lakes states (Michigan, Ohio, Wisconsin, New York, Pennsylvania, and Minnesota) have issued advisories restricting consumption of fish from some state waters due to mercury contamination. Mercury contamination, or high mercury levels in fish tissue, is also the most frequent basis for fish advisories issued by the Province of Ontario. Many of their advisories are applicable to areas of the Great Lakes.

During the early 1970s, mercury was found in fish from Lake Huron, Lake St. Clair, western Lake Erie, eastern Lake Ontario, and the St. Lawrence River at levels that led the United States and Canada to close commercial fisheries. Subsequently, mercury levels fell in these waters, because of modification or closure of certain chloralkali facilities and pulp and paper mills whose wastewater discharges contained large quantities of mercury. In 1970, mercury levels in Lake St. Clair walleye were 2 parts per million (ppm); by the mid-1980s, levels in these walleye had subsided to 0.5 ppm (Environment Canada et al. 1991). There are other signs of reduced mercury levels in the Great Lakes through dated sediment cores and populations of smelt in the lakes.

Because of large direct discharges of mercury being terminated following implementation of the Clean Water Act (CWA), the atmosphere is now the dominant pathway by which mercury reaches the Great Lakes. Currently, the best estimate of atmospheric deposition to the five Great Lakes is approximately 6,800 kg per year (15,000 pounds) (Eisenreich and Strachan 1992). Loadings of mercury to Lakes Superior and Michigan are primarily from the atmosphere. For Lake Ontario, the percentage of atmospheric contribution of mercury is relatively modest because the lake receives mercury from waters that flow from the upper lakes (Sitarz et al. 1993).

Even some of the mercury borne to the Great Lakes via their tributaries includes contamination previously deposited from the atmosphere to their watersheds.

Unlike other trace metals, mercury exists in the air predominantly in the gaseous phase due to its volatility. Estimates of wet and dry deposition of mercury to Lake Superior are about five times higher than net gas transfer to the atmosphere. The net annual atmospheric loading of mercury to Lake Superior is calculated to be about 635 kg/year (Hoff et al. 1996), which is comprised of the following estimates:

- 560 kg/year as wet deposition;
- 250 kg/year as dry deposition;
- 65 kg/year as absorption; and
- -240 kg/year as volatilization.

In an earlier study, gaseous phase mercury in the atmosphere was 1.57 ng/m^3, particulate phase mercury, 0.02 ng/m^3, and precipitation mercury, 10.5 ng/L, at a northern Wisconsin site near Lake Michigan (Fitzgerald et al. 1991). Comparison of these values with other U.S. sites is presented in Table IV-5 in Section IV.B.

In Michigan, atmospheric concentrations and wet deposition of mercury have been observed to vary geographically. Northern Michigan received only one-half the wet deposition of mercury deposited to southern portions of the state. Wet deposition varied by season, with mercury concentrations in precipitation two times greater during spring and summer than during winter. Higher levels of particulate mercury were observed in large urban areas. Modeling indicated that the dominant sources of mercury were located mostly to the south and west of Michigan (Keeler and Hoyer 1997).

Program Actions to Characterize Atmospheric Contamination in the Great Lakes

Research has occurred in the past few years to increase understanding of the effects, fate, and transport of toxic substances in the Great Lakes ecosystem. These efforts are designed to provide information to further characterize, as well as reduce, atmospheric contamination in the Great Lakes region. Some of the programs to assess the extent of atmospheric contamination in the Great Lakes basin are described below. At this time, many of these projects are collecting and/or compiling data, and results are not yet available for evaluation. Also, several notable programs/activities have been introduced in recent years to begin to reduce loadings and to mitigate existing contamination and are discussed in the following section (Toxics Reduction Efforts in the Great Lakes).

LAKE MICHIGAN MONITORING PROGRAM

A monitoring program for Lake Michigan has been implemented by EPA's Great Lakes National Program Office to support a number of activities that address reductions in the release of toxic substances, particularly persistent, bioaccumulative substances, to the Great Lakes system. The program is a key element of the Lakewide Management Plan (LaMP) for Lake Michigan (see next subsection, Toxics Reduction Efforts in the Great Lakes, for general information about the objectives of LaMPs).

The water quality criteria and values provided in the GLWQ Guidance, once adopted by the Great Lakes states, would apply to the entire Great Lakes system, regardless of the source of pollutants to those waters. In this manner, the proposed water quality criteria and the measured values provide the basis for integrating actions carried out under the range of environmental programs available to federal, state, and tribal agencies to protect and restore the Great Lakes ecosystem. The mass balance approach will facilitate this integration by evaluating multi-media load reduction actions required to ensure that Lake Michigan water quality meets the Great Lakes water quality criteria (GLWQC).

The primary goal of the Lake Michigan Monitoring Program is to develop a sound, scientific base of information to guide future toxic load reduction efforts at federal, state, tribal, and local levels. In particular, the following specific objectives are identified:

- Evaluate relative loading rates of critical pollutants by medium (atmospheric deposition, contaminated sediments, tributaries) to establish a baseline loading estimate to gauge future progress;

- Develop the predictive ability to determine the environmental benefits of specific load reduction scenarios for toxic substances and the time required to realize those benefits, including evaluation of benefits of existing environmental statutes and regulations; and

- Improve our understanding of the key environmental processes that govern the cycling and bioavailability of contaminants within relatively closed ecosystems.

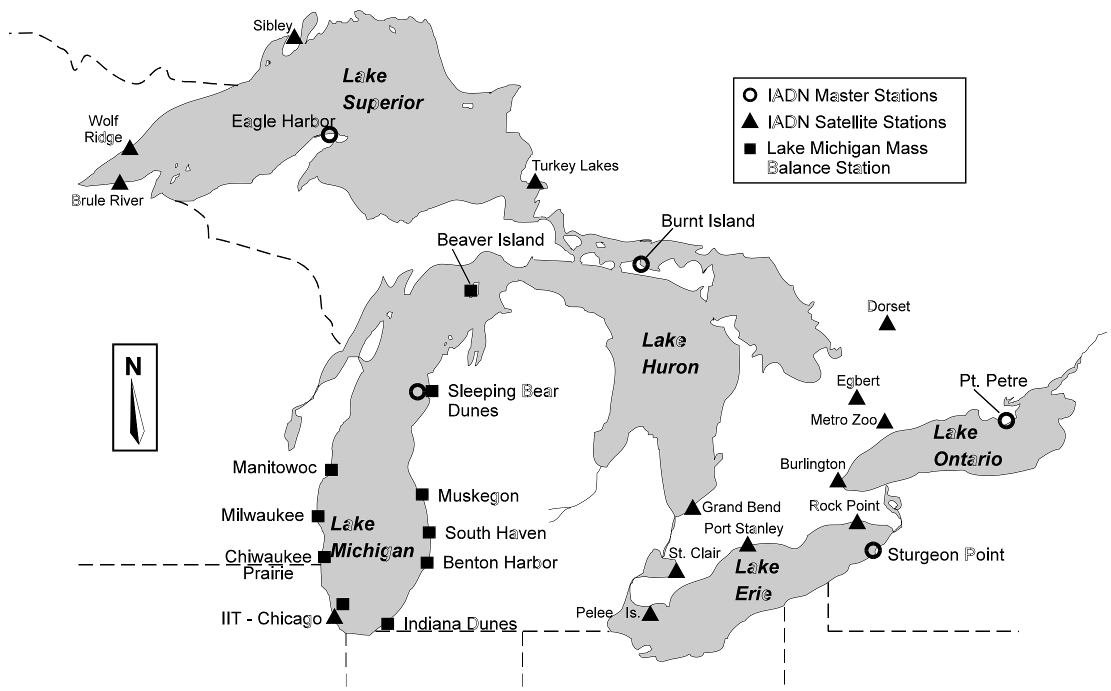
Lake Michigan Mass Balance Study. One of the ways to address the objectives of the Lake Michigan Monitoring Program, as well as to assist EPA in implementing section 112(m) of the CAA, is through a mass balance study. The mass balance study will characterize the loadings, transport, and fate of selected pollutants in a defined ecosystem, through monitoring and modeling. These measuring and estimating techniques can be applied to other ecosystems. EPA initiated the Lake Michigan Mass Balance Study, a comprehensive sampling effort that includes measurements of contaminants in the atmosphere, tributaries, lakewater, sediments, and food chain, to support model components. The atmospheric sampling sites for this mass balance study are shown in Figure IV-5.

Application of Modeling Tools from the Green Bay Mass Balance Study

In a pilot mass balance study by EPA and the Wisconsin Department of Natural Resources, water-insoluble organic compounds were monitored in Green Bay, Wisconsin, from 1988 to 1992. The analytical and modeling tools used in the study may be applied to the Great Lakes, Lake Champlain, and coastal estuaries. The **Lake Michigan Mass Balance study** is the first full-scale application of this methodology for toxic pollutants and will serve as the basis of any future mass balance efforts for persistent, bioaccumulative chemicals. Data collected for this study are anticipated in 1997.

The Lake Michigan Mass Balance model is constructed for a limited group of pollutants (PCBs, trans-nonachlor [a bioaccumulative component of chlordane], and total mercury) present in Lake Michigan at concentrations that pose a risk to aquatic and terrestrial organisms (including humans) within the ecosystem, or that may accumulate to problematic concentrations

FIGURE IV-5
Atmospheric Monitoring Sites in the Great Lakes Region



in the future and that can serve as examples for other chemicals. In addition, atrazine (under consideration for addition as a Great Waters pollutant of concern), a commonly used herbicide in the Great Lakes basin and elsewhere in the United States, is also included in the model. This herbicide has been reported at elevated concentrations in Lake Erie tributaries, in the open waters of the Great Lakes, and in the atmosphere over the lakes. The inclusion of this chemical will provide a model for the more reactive, biodegradable compounds in current use. The model will be less comprehensive than that for PCBs and trans-nonachlor, because atrazine does not appreciably bioaccumulate and it will not be analyzed in the food chain.

The chemicals chosen cover a wide range of chemical and physical properties and are representative of other classes of compounds that could pose water quality problems. This approach will allow modeling of many other chemicals with limited data. Resource limitations, quality assurance requirements, and analytical and data handling limitations preclude intensive monitoring and model calibration for more than the above described target chemicals. While mass balance modeling will focus on the above chemicals, the determination of loadings and concentrations for additional contaminants and compounds useful for source apportionment and deposition modeling will be undertaken as part of the Lake Michigan Monitoring Program.

The Lake Michigan Mass Balance Study includes an atmospheric monitoring component to address research issues concerning urban and atmospheric deposition and exchange processes. The data will be used to calculate atmospheric loads to the ecosystem. Within this air transport component, special studies are being performed to determine whether emissions of hazardous air pollutants from the urban coastal regions (Chicago, IL, and Gary, IN) contribute significantly to atmospheric deposition to the adjacent waterbodies. The objectives of these special studies are to (1) measure wet and dry deposition fluxes of urban contaminants, (2) determine contributions of urban source categories to measured concentrations and deposition rates, and (3) assess the extent of air-water exchange of contaminants.

Previous studies indicated that urban emissions have a large impact on atmospheric concentrations of air toxics and on atmospheric deposition to the Great Lakes. Dry depositional flux of PCBs from Chicago was shown previously to be three orders of magnitude higher than that of non-urban areas (Holsen et al. 1991). In addition, the Lake Michigan Urban Air Toxics Study demonstrated that concentrations of several pollutants were significantly higher in Chicago urban areas than at less urbanized sites (Keeler 1994). An intensive study was recently conducted around Chicago to assess the impact of the urban area on atmospheric deposition and exchange with Lake Michigan, with three land-based monitoring sites around Chicago and one over-water site on a research vessel approximately five miles off the shore of Chicago. The three sampling periods that occurred between 1994 and 1995 were designed to provide information to track atmospheric plumes over and across the lake. Wet deposition, dry deposition, and lake water were analyzed for semi-volatile compounds (such as PCBs and PAHs) and trace metals (such as arsenic, mercury, and lead). All samples were taken on the same day to provide information on air-water exchange of contaminants. Results from this study are expected in 1997, with modeling results expected in 1998. This study is expected to contribute useful information on urban impact to Lake Michigan, as well as to address process-oriented research issues and provide data in support of source apportionment and trajectory modeling.

INTEGRATED ATMOSPHERIC DEPOSITION NETWORK (IADN)

As mentioned earlier in this chapter, IADN is a long-term, binational program between the United States and Canada to assess the magnitude and trends of atmospheric deposition of target chemicals to the Great Lakes and to determine emission sources wherever possible. The program addresses the mandate of the Great Waters program and the needs of Annex 15 of the GLWQA between Canada and the United States. It is designed to (1) provide the necessary standardized methods, monitoring data, and loadings estimates to assess the relative importance of atmospheric deposition compared to other inputs, (2) determine temporal trends and geographic variations in deposition, and (3) ultimately provide information on sources of these atmospheric pollutants. It is a combination of a surveillance/ monitoring network and a research program. Its goals are source attribution, process identification, and assessment of atmospheric impacts on environmental systems. At this time, annual and seasonal averages have been completed for four years of IADN operation. Data for selected pollutants were presented earlier in this section. Additional details may be found in Eisenreich and Strachan (1992), Gatz et al. (1994), and Hoff et al. (1996).

Target compounds were chosen for IADN based on their potential to bioaccumulate, their tendency to be transported atmospherically, and the availability and efficiency of detection methods (see sidebar). A major benefit of IADN is the ability to monitor long-term atmospheric concentration changes of such compounds as PCBs and other chemicals of concern. In the past, such regional-scale atmospheric data have been sparse.

Compounds Measured in IADN

- ***Highest priority group:*** PCBs, lindane, PAHs, and lead. These pollutants were chosen for the first phase of IADN (1990-1992) to demonstrate the feasibility and accuracy of sampling and analytical methods.
- ***Second priority group:*** Chlorinated pesticides (such as HCB, DDT/DDE/DDD, trans-nonachlor, methoxychlor, mirex, dieldrin, aldrin) and trace metals (such as arsenic, selenium, cadmium, and mercury). Except for mercury, the sampling and analysis methods for most of these species had been implemented by 1993. Mercury monitoring was added at each IADN site by 1995.
- ***Third priority group:*** Compounds such as toxaphene, dioxins/furans, and agrochemicals which have an important atmospheric component but require additional methods development to accurately measure their concentrations in atmospheric deposition samples.

It was originally projected that, to be representative of regional deposition patterns, the IADN required one "master" station and several "satellite" stations on each lake. The master stations are: Eagle Harbor, MI; Sleeping Bear Dunes, MI; Burnt Island, Ontario; Sturgeon Point, NY; and Pt. Petre, Ontario (see Figure IV-5). Several satellite sites were later added, including an urban site in Chicago (see box on next page). The IADN implementation design allows for periodic evaluation of the existing sites to determine whether other sites are needed.

At each IADN site, concentrations of target chemicals are measured in rain and snow (wet deposition), airborne particles (dry deposition), and airborne organic vapors. In addition, precipitation rate, temperature, relative humidity, wind speed and direction, and solar radiation are measured at each site. IADN results for selected pollutants are presented in Tables IV-1 and IV-2.

At this time, after five years of operation, many of the sampling and analysis issues of IADN have been resolved. For example, comparability of sampling and analytical procedures between jurisdictions was achieved through extensive laboratory intercomparison studies. However, the uncertainty in the analytical measurement of some compounds is still above the uncertainty threshold acceptable to most policy makers. Toxic chemicals at extremely low concentrations in air, such as PCBs and some agricultural chemicals, have the highest uncertainties in sampling (over 40 percent). Relative standard deviations of air concentrations of organochlorinated compounds may vary from 60 to 90 percent, due to seasonal and annual fluctuations in the air, rather than precision of the measurement (Hoff et al. 1996).

Uncertainty in the deposition estimates may result from various factors: (1) general approximations for estimating deposition; (2) climatic and meteorological variations; (3) differences in the instrumentation and the scope and objectives of the various jurisdictions and agencies involved; and (4) estimation of factors used to calculate loadings (e.g., magnitude of Henry's Law constants, rates of contaminant transfer between the air and water). Despite these

Urban Influence on Atmospheric Deposition of Contaminants

The primary focus of IADN is to determine regionally representative atmospheric deposition loadings of toxic chemicals to the Great Lakes. Thus, monitoring stations were positioned to minimize the influence of local sources and to monitor the atmospheric environment over the lakes as much as possible. This approach does not directly enable the determination of the role of urban air pollution. Recent research suggests that deposition of contaminated large particles carried by winds passing over urban areas can result in substantial inputs of toxic chemicals to the Great Lakes (Falconer et al. 1995; Holsen et al. 1991).

The influence of pollution from the Chicago-northwest Indiana area on water quality in southern Lake Michigan was studied by Sweet and Basu (1994). The Sleeping Bear Dunes site (in the State of Michigan) is located one kilometer from the northeastern shore of Lake Michigan and 50 kilometers from the nearest urban area or major source and, thus, is considered a "remote" site. The first urban site is located 1.5 kilometers from the shore on the campus of the Illinois Institute of Technology, which is near major expressways and surrounded by commercial and residential areas. The second urban site is located at the Indiana Dunes National Lakeshore in the vicinity of large steel mills. Particulate concentrations were measured for target compounds (PCBs, pesticides, and trace metals). Gas concentrations of PCBs and pesticides were determined, and rain was analyzed only for PCBs.

Results from Sweet and Basu (1994) indicate that, for PCBs, DDT (and its metabolites), dieldrin, chlordane, and several trace metals (manganese, zinc, chromium, and lead), the measured particulate and gas concentration values were 10 to 40 times higher in urban areas than at the remote site. For other pesticides (α-HCH, lindane, HCB) and trace metals (arsenic and selenium), concentrations were nearly the same at all three sites, indicating these pollutants were well mixed in the air throughout the region (and that there were probably few local sources).

Though 90 to 99 percent of the PCBs were found in the gas phase, the most toxic PCB congeners were enriched in the particulate phase (Falconer et al. 1995; Holsen et al. 1991; Sweet and Basu 1994). Thus, dry deposition may be an important transport mechanism for certain, especially toxic, PCBs to the lakes. Urban particulate matter also carried high concentrations of trace metals and pesticides, causing dry deposition of these materials to southern Lake Michigan. Dry deposition of large particles may be especially significant for Lake Michigan because 200 kilometers of the southwest shoreline are heavily developed. Prevailing southwest winds carry emissions over the lake where they travel for 100 to 150 kilometers before reaching land again, allowing a significant portion of the deposition to enter the lake. Finally, the concentration of PCBs in precipitation is roughly the same in urban and rural sites. The relatively low levels found in urban precipitation may be due to the fact that many contaminants are collected in clouds or by rain upwind of polluted areas.

Clearly the influence of urban areas on atmospheric deposition of certain pollutants to the Great Lakes is substantial, especially in heavily developed areas, such as the southwestern shores of Lake Michigan.

limitations, the reported estimates are the best that are currently available. Also, data on the concentration of contaminants in the water column for all the Great Lakes have improved recently with more samples being collected and analyzed.

GREAT LAKES EMISSIONS INVENTORIES

A significant step toward assessing the need to reduce atmospheric loads of hazardous air pollutants to the Great Lakes is to identify, categorize, and estimate the magnitude of the pollutant sources. By creating an emissions inventory data base, it is possible to identify the sources and source categories that contribute most to the total emissions in a given geographic area, as well as to model emissions transport and deposition. An air emissions inventory is typically based on mathematical estimates of pollutant releases through the use of emission factors (e.g., a number that represents emissions per unit burned, produced, or processed). These emission factors are derived from actual measurements of the emissions from representative sources and are derived specifically for one type of process or process equipment. Emission factors can be used, for example, to estimate both the amount and type of pollutants being emitted from an air pollution source based upon the quantities of material processed.

The 1986 Great Lakes Governors' Toxic Substances Control Agreement specified provisions to address atmospheric deposition, including a commitment "to cooperate in quantifying the loadings of toxic substances originating from all sources, with the purpose of developing the most environmentally and economically sound control programs." In response to the governors' direction, the air regulatory agencies in the eight Great Lakes states and the province of Ontario began to work cooperatively in 1987 to investigate "the development of a computerized air toxics data base for the purpose of obtaining a better understanding of the nature and sources of toxic air emissions and their migration, dispersion, and resulting impact upon the Great Lakes basin." Under the auspices and management of the Great Lakes Commission (representing the eight Great Lakes states) and with major funding from EPA's Great Waters program, the Great Lakes states began developing a *regional* air toxics emissions inventory. This first regional inventory is scheduled for completion in 1997 and is expected to compile 1993 emissions data for 49 toxic air pollutants from point and area sources. Emissions data on toxic air pollutants from mobile sources will be developed in 1997-1998. These 49 pollutants include 10 of the Great Waters pollutants of concern (cadmium, chlordane, hexachlorobenzene, lead/alkylated lead, mercury, PCBs, PAHs, POMs, TCDD, TCDF). The continued partnership of the region's air regulatory agencies, now in its eighth year, and the high level of regional cooperation and coordination exemplifies the commitment to decreasing toxic deposition into the Great Lakes ecosystem. Yet the inventory must be accompanied by an ongoing commitment to further quantify, assess, and report on the effects of voluntary and regulatory reductions of air toxics emissions.

The key to the state's coordinated efforts is *The Air Toxics Emissions Inventory Protocol for the Great Lakes States*, developed in June 1994 (see sidebar). The Protocol will be followed by each participating state ensuring that consistent, agreed-upon best methodologies are used among all states when compiling a quality-assured inventory. This Protocol is an evolving document and will be updated or revised as needed and agreed upon by all the Great Lakes states.

The second fundamental component in developing a toxic air emissions inventory is the Regional Air Pollutant Inventory Development System (RAPIDS), a multi-state pollutant emissions estimation and storage software system. RAPIDS is a state-of-the-art, networked, relational data management and emission estimation

Components of Great Lakes Emissions Inventory

To date, the Great Lakes States have developed and tested two fundamental components of the inventory effort:

1. ***The Air Toxics Emissions Inventory Protocol for the Great Lakes*** - A guide for each state's efforts to identify sources and estimate emissions so that the inventory is complete, accurate, and consistent from state to state.

2. **RAPIDS** - A client/server relational database software and data management and emissions estimation system. It was designed so that the Great Lakes states may adopt RAPIDS (or some variant of it) for their state system, and may also submit their data for incorporation with the regional RAPIDS data base at EPA.

system, bridging each state's individual inventory and computer system to the regional RAPIDS repository of inventory data. RAPIDS' strength is its versatility. States can modify or build upon it to serve their particular needs. It can be used to estimate both toxic and criteria pollutant emissions from a single device within a facility or a complex grouping of devices and controls, or even across geographic areas, ranging in size from one facility to the entire Great Lakes region. It is designed to run on a personal computer and applies a flexible data model that can be easily expanded in the future to support multi-media, permitting, monitoring, reporting, and compliance activities in the states. Emission factors are uploaded from EPA's Factor Information Retrieval System (FIRE), which contains quality-rated emission factors for both criteria and hazardous air pollutants.

Using RAPIDS, the Great Lakes states' air regulatory agencies are building a comprehensive, updatable statewide and regional air toxics inventory for point, area, and (in the future) mobile sources for the 49 air pollutants. Each of the eight Great Lakes states will be responsible for compiling, uploading, and validating their state emissions inventory data. It is anticipated that the inventories will be updated on a one- or two-year basis.

Four states (Illinois, Indiana, Wisconsin, and Michigan) completed a pilot study of major urban areas along the southwest shore of Lake Michigan in December 1995 using RAPIDS and the Protocol. The states created an inventory of small point and area source categories in the 12 shoreline counties encompassing Chicago (Illinois), Gary (Indiana), and Milwaukee (Wisconsin) that contribute the most to the total emissions of the 49 pollutants of concern. These area sources include gasoline stations, foundries, asphalt and cement plants, and hospitals, among others. The project was the first rigorous test of the regional Protocol and the RAPIDS software. Total pollutant emissions from the inventoried sources were collected, but data interpretations and conclusions were not developed from the results. Instead, the process of compiling the regional inventory was used as a means of resolving many technical, methodological, and policy-related issues that impact a multistate, regional toxic air emissions inventory. Furthermore, the pilot study provided useful information on serious shortcomings that still exist in the regional emissions estimates and suggested necessary steps that must be made to ensure data quality for estimating various pollutant groups. The results from this pilot study will also contribute for better methodology for use in the full eight-state regional inventory.

The level of emissions resolution and the source categories contained in RAPIDS were planned to meet the modeling needs of Great Lakes air quality researchers. This inventory will be available for dispersion and deposition models to characterize source, source category, and geographic contributions, and for mass balance models to characterize media contributions.

Toxics Reduction Efforts in the Great Lakes

In recent years, several programs/activities have developed approaches to reduce loadings and to mitigate existing contamination. These programs are described below. They may provide information to further characterize and reduce atmospheric contamination in the Great Lakes region.

VIRTUAL ELIMINATION

The Great Lakes Water Quality Agreement of 1978 between the United States and Canada called for the "virtual elimination" of persistent toxic substances, especially those which bioaccumulate, from the Great Lakes basin. In keeping with the obligations of the Great Lakes Water Quality Agreement, two major efforts have occurred: (1) a pilot project sponsored by EPA to develop the framework to achieve virtual elimination of two pollutants, mercury and PCBs; and (2) development of the Great Lakes Binational Toxics Strategy (released in April 1997) between the United States and Canada to set goals to reduce the use and release of selected pollutants. Many of the recommendations from the pilot project were incorporated into the Binational Toxics Strategy.

Virtual Elimination Pilot Project. Since 1990, both the United States and Canada have initiated separate efforts for virtual elimination. EPA began the Virtual Elimination Pilot Project in 1993, which was designed to answer the following question: "What options exist for improving the current regulatory and non-regulatory framework to encourage continuing reductions towards zero in the use, generation, and release of selected toxic substances?" The aim of the project was to identify barriers to achieving virtual elimination and to develop strategies to overcome these barriers.

The first iteration or "pilot" portion of the U.S. project focused on the reduction opportunities for two substances, mercury and PCBs. EPA held a meeting with stakeholders in the Great Lakes region in 1993 to share information on mercury and PCBs, and to offer participants the opportunity to make recommendations on ways to reduce the use and release of each pollutant. Based on the results of this meeting, a draft report was developed by EPA to identify options to reduce mercury (GLNPO 1995). A draft options paper for the virtual elimination of PCBs is currently being prepared by EPA. It is expected that this project will continue with the analyses of classes of substances rather than the use of a chemical-by-chemical approach.

Mercury presents an unusual challenge to society because of its semi-volatility, persistence, complex environmental chemistry, and tendency to bioaccumulate in fish. The draft mercury options paper (GLNPO 1995) proposed regulatory and voluntary measures to prevent or reduce atmospheric mercury contamination, and introduced the concept of the mercury "life cycle." A comprehensive approach to virtually eliminating mercury releases was proposed:

- Increase public awareness of mercury problems and mercury-containing items;
- Influence supply of mercury to minimize primary production and manage federal holdings;
- Minimize use of mercury through pollution prevention and alternative technologies;
- Reduce uncontrolled releases by encouraging recycling and regulating releases; and
- Manage disposal of mercury-containing items and mercuric wastes.

Regulatory and Voluntary Options that may Prevent or Reduce Atmospheric Mercury Contamination

- ***Pollution Prevention.*** Mercury use in the United States has substantially declined during the past two decades. This trend parallels that of western Europe. Ebbing use implies that less mercury will be included in waste streams reaching incinerators and landfills, or released during production processes. The decline in use is attributable to both government bans and technological advances that create environmentally safer products. Mercury has been banned in pesticides (1972-1976 and 1993) and in paints. The number of U.S. chloralkali facilities using a mercury cell process has declined from 25 during the early 1970s to 14 in recent years. Minnesota, Wisconsin, and New York restricted mercury content in batteries, and consequently, national use of mercury in batteries fell from 448 tons during 1988 to 10 tons during 1993. Mercury content in fluorescent bulbs has also declined.

- ***Recycling.*** A number of users of mercury are taking steps to boost recycling. Various municipal wastewater treatment plants are working with dental associations to encourage recovery of mercury in dentist offices so that mercury amalgam does not enter their treatment systems. Some manufacturers who rely on mercury in instruments are starting or considering take-back programs; EPA and states are working with several such firms to ensure that liability concerns do not preclude voluntary efforts. A number of hospitals are diverting mercury-containing wastes from incineration. Several states require that fluorescent bulbs be recycled (e.g., Florida, Minnesota), because such bulbs are commonly broken in or en route to landfills, allowing mercury vapors to escape to the atmosphere. While the quantity of mercury contained in a single bulb is minuscule, the number of discarded bulbs is great.

- ***Management.*** Pursuant to implementation of the CAA, EPA has proposed performance standards for municipal and medical waste incinerators. A final rule for municipal waste combustors (new sources with individual capacity of >250 tons/day) was promulgated in December 1995 (60 *Federal Register* 65387) and a proposed rule for medical waste incinerators in June 1996 (61 *Federal Register* 31736). There are about 2,400 medical and 180 municipal incinerators across the nation. These sectors are estimated to generate about one-half of current national emissions of mercury, primarily through combustion of discarded products that contain mercury. When fully implemented by 2002 through state plans, these proposed standards hold promise of reducing mercury emissions from these two classes of incinerators by 95 and 80 percent, respectively. Performance standards will also be evaluated for other sectors that also emit mercury, including the Portland Cement industry; commercial and industrial boilers; primary lead smelters; the chloralkali sector; primary copper smelters; sewage sludge incinerators; and lime manufacturing.

- ***Global Production and Release of Mercury.*** During recent years, the U.S. government has held a significant stockpile of mercury on the world commodity market. To dispose of its holding, the Department of Defense has periodically auctioned mercury. From 1988 through 1993, sales totaled four million pounds. Sales were suspended in 1994, pending consideration of their environmental impacts. One positive environmental impact of sales is that they may forestall virgin production of mercury; the last mercury mines in the United States closed several years ago. Both western Europe and the United States have become substantial net exporters as their mercury consumption has fallen, whereas world use may be growing (Lawrence 1994). Because of diminished use of mercury, the United States is nearly meeting its entire need for mercury through recycling. Much of the mercury sold by the federal government has been exported. Foreign use and release, due to less stringent controls, can contribute to global atmospheric contamination which can travel for long distances, and directly contaminate U.S. surface waters. Suspension of government sales has positioned the United States to seek needed international cooperation in minimizing mercury releases on a global scale.

The Great Lakes Binational Toxics Strategy. The *Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes*, also known as the Binational Great Lakes Toxics Strategy, was signed between the two countries on April 7, 1997 (U.S. EPA and Environment Canada 1997). This Binational Strategy was developed jointly by EPA and Environment Canada, in keeping with the objectives of the 1987 GLWQA. Both Canada and

United States have domestic virtual elimination strategies but a coordinated strategy is necessary for the greatest reduction in toxic substances throughout the Basin. Both nations encourage and support voluntary programs by all stakeholders to reduce the generation, use, and release of toxic substances to the Great Lakes.

The Binational Strategy provides the framework to achieve quantifiable goals in a specified time frame (1997 to 2006) for targeted persistent toxic substances, especially those which bioaccumulate. Flexibility is provided in the Strategy to allow for the revision of targets, time frames, and the list of pollutants. All actions and activities, both regulatory and nonregulatory, will be considered to help speed reductions. The Strategy also recommends that goals be accomplished through a four-step process (see sidebar). The pollutants identified in the Strategy fall into two lists. The Level I substances represent an immediate priority and are targeted for reduction and eventual virtual elimination through pollution prevention and other incentive-based actions. These pollutants are aldrin/dieldrin, benzo(a)pyrene, chlordane, DDT/DDE/DDE, hexachlorobenzene, alkyl lead, mercury and mercury compounds, mirex, octachlorostyrene, PCBs, dioxins/furans, and toxaphene. For pollutants that are considered Level II substances, the governments encourage stakeholders to undertake pollution prevention activities to reduce levels in the environment of those substances nominated jointly by both countries, and to conform with the laws and policies of each country, including pollution prevention, with respect to those substances nominated by only one country, until and unless these pollutants are placed on the Level I list. The Level II pollutants are cadmium and cadmium compounds, 1,4-dichlorobenzene, 3,3'-dichlorobenzidine, dinitropyrene, endrin, heptachlor (and heptachlor epoxide), hexachlorobutadiene (and hexachloro-1,3-butadiene), hexachlorocyclohexane, 4,4'-methylenebis(2-chloroaniline), PAHs, pentachlorobenzene, pentachlorophenol, tetrachlorobenzene (1,2,3,4- and 1,2,4,5-), and tributyl tin.

A Four-Step Process Toward Virtual Elimination

1. Gather information on generation, uses, and sources of the pollutant within and outside the Great Lakes Basin;

2. Analyze current regulatory and non-regulatory programs and initiatives that manage or control the pollutants and identify the gaps in these regulations that offer opportunities for reductions;

3. Develop cost-effective options and provide recommendations for increasing the pace and level of reductions; and

4. Recommend and implement actions to achieve goal.

Both the United States and Canada have set "challenge" goals to achieve reductions through implementation of voluntary efforts and regulatory actions. One of these challenges is the commitment of these countries to work together to assess atmospheric inputs of persistent toxic substances to the Great Lakes, with the goal of evaluating and reporting jointly on the contribution and significance of long-range transport of these substances from worldwide sources. Efforts will be made to work within the existing international framework to reduce releases of such pollutants from remaining long-range sources. Activities by EPA and Environment Canada to meet this particular challenge include:

- Coordinate efforts to identify sources in order to better define and coordinate emission control programs;

- Maintain atmospheric deposition monitoring stations to detect deposition and transport of toxic substances;

- Continue research on atmospheric science of toxic pollutants to refine and to improve existing source, receptor, and deposition models, as well as improve integration of existing air toxic monitoring networks and data management systems to track deposition of contaminants within the Great Lakes; and

- Conduct an assessment of long-range transport of persistent toxic pollutants from worldwide sources

In addition to these coordinated binational efforts, Environment Canada will also demonstrate alternative processes to lessen emissions from five predominant sources by 2001 and complete inventories of 10 selected air pollution sources to support assessment of environmental impacts of air toxics by 1999.

Besides the above challenge, the Strategy includes several specific reduction goals or challenges for the Level I pollutants (Table IV-3). For the United States, the baseline from which these reductions will be measured will be the most recent and appropriate inventory (e.g., mercury will be based on estimated emissions during the early 1990s). Canada plans to use their 1988 emissions inventory.

Two additional challenges from the Strategy are: (1) complete or be well advanced in remediation of priority sites with contaminated bottom sediments in the Great Lakes Basin by 2006; and (2) promote pollution prevention and sound management of Level II substances, to reduce levels in the environment. The Binational Strategy is intended to fill in the gaps that exist where ongoing programs or emerging initiatives do not address toxic releases, to provide a context of basinwide goals for localized actions, and to provide "out of basin" support to programs such as LaMPs.

LAKEWIDE MANAGEMENT PLANS (LAMPS)

In Article VI, Annex 2 of the GLWQA, the U.S. and Canadian governments agreed to develop and implement LaMPs for each of the five Great Lakes. The purpose of the LaMPs is to document an approach to reducing input of critical pollutants to the Great Lakes and restoring and maintaining Great Lakes integrity. LaMPs are management tools designed to (1) integrate federal, state, provincial, and local programs to reduce loadings of toxic substances from both point and nonpoint sources; (2) assess whether these programs will ensure attainment of water quality standards and designated beneficial uses; and (3) recommend any media-specific program actions or enhancements to reduce toxic loadings in waters currently not attaining water quality standards and/or designated beneficial uses. Unlike the other four Lakes, Lake Michigan lies entirely within the boundaries of the United States and therefore, the Lake Michigan LaMP has been developed solely by U.S. federal and state agencies with input from a public forum. The development of this program, as well as the deadlines established for the completion of the program, is mandated under section 118 of the CWA. In addition, as noted in Chapter I, section 112(m) of the CAA requires that EPA, in cooperation with NOAA, monitor the Great Lakes, investigate atmospheric deposition rates and pollutant sources, improve monitoring methods, and determine the relative contribution of atmospheric pollutants to the total pollution loadings to the Great Lakes and other Great Waters.

TABLE IV-3
Specific Pollutant Reduction Goals Under the Great Lakes Binational Toxics Strategy [a]

Level I Substances	United States Challenge	Canadian Challenge
Aldrin/Dieldrin, Chlordane, DDT, Toxaphene, Mirex, Octachlorostyrene	Confirm by 1998 that there is no longer use or release from sources that enter Great Lakes Basin. If ongoing long-range sources from outside of U.S. are confirmed, use existing international frameworks to reduce or phase out releases.	Report by 1997 that there is no longer use, generation, or release from Ontario sources that enter Great Lakes Basin. If ongoing long-range sources outside of Canada are confirmed, use existing international frameworks to reduce or phase out releases.
Alkyl lead	Confirm by 1998, there is no longer use in automotive gasoline; support and encourage stakeholder efforts to reduce releases from other sources.	Seek by 2000, 90 percent reduction in use, generation, or release.
PCBs	Seek by 2006, a 90 percent reduction nationally of high level PCBs (>500 ppm) in electrical equipment; ensure all PCBs retired from use are properly managed and disposed of to prevent accidental releases within or to the Great Lakes Basin.	Seek by 2000, a 90 percent reduction nationally of high-level PCBs (>1 percent PCBs) that were once, or are currently, in service and accelerate destruction of stored high-level PCB wastes that may enter the Great Lakes Basin.
Mercury[b]	Seek by 2006, a 50 percent reduction nationally in deliberate use of mercury and a 50 percent reduction in release from sources resulting from human activity.[c]	Seek by 2000, a 90 percent reduction nationally in releases of mercury, or where warranted the use of mercury, from polluting sources resulting from human activity in the Great Lakes Basin.
Dioxins/Furans	Seek by 2006, a 75 percent reduction in total releases of dioxins/furans (2,3,7,8-TCDD toxicity equivalents) from sources resulting from human activity.[c]	Seek by 2000, a 90 percent reduction in releases of dioxins/furans (focus on 2,3,7,8-substitute congeners) from sources resulting from human activity in Great Lakes Basin.
Benzo(a)pyrene, HCB	Seek by 2006, reductions in releases that are within or may have potential to enter the Great Lakes Basin from sources resulting from human activity.	Seek by 2000, a 90 percent reduction in releases from sources resulting from human activity in the Great Lakes Basin.

[a] Detailed descriptions of these challenges are presented in The Great Lakes Binational Toxics Strategy (U.S. EPA and Environment Canada 1997).
[b] Mercury challenges are considered interim reduction targets for mercury and, in consultation with stakeholders, will be revised if warranted, following completion of EPA's Mercury Study Report to Congress (U.S. challenge) and 1997 *Canada-Ontario Agreement Respecting the Great Lakes Basin Ecosystem* (Canadian challenge).
[c] The release challenge applies to the aggregate of releases to the air nationwide and of releases to the water within the Great Lakes Basin.

A LaMP is a dynamic, action-oriented process encompassing a number of components. These include an evaluation of beneficial use impairments and pollutants contributing to those impairments; a summary of sources and loads of these critical pollutants; identification of ongoing prevention, control, and remediation actions, as well as additional efforts needed to reduce pollutant loads and to restore beneficial uses; and monitoring activities to evaluate the effectiveness of program actions. This approach for developing and implementing LaMPs is an evolutionary and iterative process for identifying and reducing critical pollutants. Public participation and cooperation with states and local governments is a key component to the LaMP development process.

LaMPs are in various stages of development for each of the Great Lakes (see sidebar). Not all of the Lakes have LaMPs published in the Federal Register; however, commitments have been made by key stakeholders in the respective basins to pursue toxics reduction. Actions are being taken to achieve this goal. Each LaMP addresses a different list of critical pollutants, but some common ones are mercury, PCBs, hexachlorobenzene, dioxins, furans, chlordane, DDT and metabolites, and dieldrin (all of which are Great Waters pollutants of concern).

Current Status of LaMPs in the Great Lakes

Superior	Binational Program to Restore and Protect the Lake Superior Basin announced (1991) Stage 1 LaMP submitted to IJC (1995) Stage 2 LaMP released for public review (1996)
Michigan	LaMP published in Federal Register (1994)
Huron	LaMP not established
Erie	LaMP Management Committee formed (1994)
Ontario	Lake Ontario Toxics Management Plan (1989) LaMP Workplan signed (1993)

Several activities have been initiated through LaMPs:

- In the Lake Michigan basin, agricultural "clean sweeps" to properly collect and dispose of unused pesticides have been conducted in Indiana, Michigan, and Wisconsin. Also, a variety of pollution prevention and technical assistance projects have taken place in Milwaukee, Chicago, and western Michigan.

- In most lake basins, tributary and atmospheric deposition monitoring is occurring. The Lake Michigan LaMP is utilizing the information generated from the Lake Michigan Mass Balance Study (described earlier in the section) to identify and reduce loadings as data become available.

- The Lake Superior LaMP was initiated as a component of the binational efforts to restore and protect Lake Superior. One of the goals of the LaMP is to achieve zero discharge and emission of persistent toxic pollutants through its Zero Discharge Demonstration Project. Another effort of the Lake Superior LaMP is an extensive pollution prevention outreach and education program developed for mercury. Among the activities are battery collection, energy efficiency, and product takeback programs. For example, Honeywell, Inc., the largest manufacturer of mercury thermostats used in regulating heating in the home, has instituted a thermostat takeback program in which the company recycles the mercury.

The LaMPs often provide the needed coordination and oversight for many such projects being implemented all over the Great Lakes Basin.

THE GREAT LAKES WATER QUALITY (GLWQ) GUIDANCE

Another notable toxics reduction effort was the GLWQ Guidance (U.S. EPA 1995a). It is the culmination of a six-year cooperative effort that included participation by the eight Great

Lakes states, the environmental community, academia, industry, municipalities, and EPA's regional and national offices, and stems from the Great Lakes Water Quality Initiative, which began when the states of the Great Lakes Region recognized the important feature of the Great Lakes ecosystem to accumulate persistent pollutants. The guidance is not only designed to address existing problems, but also to prevent emerging and potential problems posed by additional chemicals in the future, which may damage the overall health of the Great Lakes system. The guidance includes criteria for the protection of human life, wildlife, and aquatic life, taking into account the ability of many pollutants to biomagnify. Antidegradation requirements assure that current water quality will not be diminished. The guidance also outlines procedures to ensure consistent implementation and appropriate flexibility for long-term protection of the Great Lakes.

The GLWQ Guidance promotes the use of pollutant minimization plans to stop pollution before it enters the environment. Reducing pollution at its source is the most effective way of protecting public health and the environment, and is often more economical than cleaning up after a pollutant is released.

The water quality criteria in the GLWQ Guidance apply to all of the waters in the Great Lakes system, regardless of the source of pollution. Pollutants enter the Great Lakes from the air, stirred-up bottom sediments, urban and agricultural runoff, hazardous waste and Superfund sites, spills, and industrial and municipal wastewater discharges. Although the implementation procedures of the guidance apply mostly to industrial and municipal water discharges, a state may find it is more effective (or cost-effective) to improve water quality by reducing air emissions or cleaning up contaminated sediments, and may choose not to impose additional requirements on wastewater dischargers.

GREAT LAKES REMEDIAL ACTION PLANS

The 1978 GLWQA, along with the 1987 amendments, established guidelines for restoring the quality of the Great Lakes. As a response to this measure, geographical "problem areas" or Areas of Concern (AOCs) were identified in the Great Lakes where GLWQA objectives had been exceeded and such exceedance had caused, or was likely to cause, impairment of beneficial use or the area's ability to support aquatic life. The defined AOCs include rivers, connecting channels, harbors, and embayments of the Great Lakes, with the U.S. states and Canadian provinces responsible for remediating these areas. The Water Quality Board (WQB) of the IJC determines the AOCs, but the specific geographical boundaries of the AOCs are set by the states and/or provincial governments. Currently, there are 43 AOCs; 12 are under Canadian jurisdiction, 26 under U.S. jurisdiction, and five under jurisdiction of both countries. The sources of contamination have usually been water discharges from point and non-point sources.

In order to provide more uniform guidance on how to remediate the AOCs, Remedial Action Plans (RAPs) were introduced in 1985. In 1994, eight U.S. states were involved in the RAP process: Indiana, Illinois, Minnesota and Pennsylvania (1 AOC each), Ohio (4 AOCs), Wisconsin (5 AOCs), New York (6 AOCs), and Michigan (14 AOCs). Currently, for most AOCs, the problem definition stage of the RAP process has been addressed and the planning and implementation stages are to be initiated. One of the major problems facing the AOCs today is toxic contamination of the sediments. All of the U.S. AOCs have impaired beneficial uses attributable to contaminated sediments (U.S. EPA and Environment Canada 1994). As a result, sediment remediation is a key component of RAP remediation.

The progress of AOC remediation is presented in an EPA report, *Progress in the Great Lakes Remedial Action Plans: Implementing the Ecosystem Approach in the Great Lakes Areas of Concern* (U.S. EPA and Environment Canada 1994). This report provides an update for each AOC in the Great Lakes and summarizes the major barriers to and benefits of the RAP process. It concludes that a comprehensive decision-making process that leads to commitment for action is an essential aspect of implementing an ecosystem approach, as required in a RAP. Great Lakes federal, state, and provincial governments have provided leadership and resources for development and implementation of RAPs. Through government and community-based partnerships, RAPs are being developed to be a coordinated, multi-stakeholder response to restoring impaired beneficial uses in AOCs.

SOME ADDITIONAL ACTIONS RELATED TO TOXIC CONTAMINATION AND REDUCTION

In addition to activities described above, many pollutant-specific efforts provide significant information on atmospheric contamination in the Great Lakes. For example, EPA developed standards for municipal waste combustors (excludes new sources with individual capacity of $\leq$250 tons/year) (60 *Federal Register* 65387) and proposed standards for medical waste incinerators (61 *Federal Register* 31736) which will, when implemented by 2002, provide about a 70 ton reduction in mercury emissions, or 35 percent of current total U.S. emissions, based on 1990 emissions estimates. Implementation of other Maximum Available Control Technology (MACT) standards, including those proposed in 1996 for hazardous waste combustors, offer the probability of further mercury emission reductions in the future.

For toxaphene, many issues remain about its sources and continuing presence in the Great Lakes. EPA held a research workshop on March 27-29, 1996, with scientists to assess these concerns (U.S. EPA 1996c). Based on the meeting, the following actions were recommended to maintain progress toward resolving several questions concerning concentrations and trends (spatial and temporal) in various Great Lakes media and the sources of toxaphene in the Great Lakes:

- Measure concentration gradient of toxaphene across air-water interface and link with atmospheric source profile to establish seasonal and annual fluxes to and from Lakes Superior and Michigan, as well as correlative measurements in other Great lakes to permit spatial comparisons;

- Collect additional sediment cores in Lakes Superior and Michigan to determine concentrations, accumulation rates, and inventories to help establish past dynamics of toxaphene and assist in efforts to forecast future conditions in the Great Lakes;

- Quantify aquatic food web dynamics to establish how food web influences spatial and temporal variations in toxaphene concentrations in biota.

- Measure physical-chemical properties of toxaphene homologs and congeners.

Besides providing information on sediment deposition and cycling of toxaphene in the Great Lakes, it is anticipated that results from these recommendations would provide a more firm technical basis upon which to explore the need for and extent of appropriate management actions.

Another major binational effort to broadly address Great Lakes issues is the State of the Lakes Ecosystem Conference (SOLEC) organized by the governments of the United States and Canada. In the first SOLEC in 1994, a report (and five background papers) was released on the current condition of the Great Lakes (Environment Canada and U.S. EPA 1995). The report addressed the entire Great Lakes system in terms of ecological and human health, and the stressors which affect it. The six areas of discussion were human health; aquatic community health; aquatic habitat; toxic contaminants; nutrients; and the economy. Although the report did not describe or evaluate pollution control or natural resource management programs, it did focus on environmental conditions within the Great Lakes system. The SOLEC report indicated that there has been considerable improvement in all the Great Lakes compared to 30 years ago, although serious losses in habitat for native plants and animals continues. Nutrient and toxic contaminant concentrations appear to be decreasing, although bioaccumulative pollutants still cause problems. The report presented a "mixed picture" of the current conditions of the Great Lakes and challenges managers and decision makers throughout the basin to obtain adequate information, deal with subtle effects of long-term exposure to low levels of toxic contaminants, protect biodiversity, restore habitat for native plants and animals, connect decisions with ecosystem results, and attain sustainability.

State of the Lakes Ecosystem Conference (SOLEC)

As part of the continuing response by the governments of the United States and Canada to the binational Great Lakes Water Agreement, SOLEC was initiated and held in October 1994. The second meeting of SOLEC was held in Windsor, Ontario, on November 6-8, 1996. SOLEC is viewed as part of a process of sharing information needed to make well informed decisions that affect the ecosystem of the Great Lakes. It is attended by managers and other decision makers from the private sector, and government and non-government environmental organizations.

Building on the findings of the first SOLEC, the 1996 SOLEC (Environment Canada and U.S. EPA 1996) focused on nearshore areas of the Great Lakes basin (i.e., warm and shallow waters, coastal wetlands). These areas represent the most diverse and productive parts of the Great Lakes ecosystem, and provide support for most intense human activity and subsequently, are subjected to greatest stress. Key themes from the conference were immediate actions; local/community level involvement; development of common ecosystem health indicators to measure progress; essentiality of cooperation and partnership due to complexity of issues and development of new ideas; approaches that recognize long-term perspective; and focus on prevention and preservation. The Great Waters program will benefit from many aspects of SOLEC, such as the information gathered on the impact of air pollutants on human health and ecological effects and the promotion of pollution prevention measures.

Addressing Data Gaps/Future Needs

Considerable progress has been made in the recent past in characterizing and reducing toxic pollution in the Great Lakes. The programs presented above and summarized in Table IV-4 provide an overview of some notable and recent activities by the United States, as well as Canada, to respond to concerns related to atmospheric pollution in the Great Lakes. Some of these measures include identifying emission sources; characterizing contamination from

TABLE IV-4
Summary of Some Major Programs to Address Atmospheric Contamination in the Great Lakes

Data Collection/Research Projects	
Lake Michigan Monitoring Program/Lake Michigan Mass Balance Study	Scientific base for future load reduction effort at all government levels. Mass balance study addresses CAA section 112(m) through coordinated effort to quantify and understand loadings, transport, and fate of selected HAPs. Also to provide a validated method to estimate loading for other waterbodies.
Integrated Atmospheric Deposition Network (IADN)	Binational monitoring network and research program to determine magnitude and trends of atmospheric deposition for the region.
Great Lakes Emissions Inventories	Inventory of sources and source category emissions in Great Lakes region, with a multi-state data base (RAPIDS).
Toxics Reduction Efforts	
Virtual Elimination Pilot Program/ Great Lakes Binational Toxics Strategy	Regulatory and non-regulatory efforts to encourage reduction in use and release of bioaccumulative pollutants in Great Lakes.
Lakewide Management Plan (LaMP)	Management tool to document approach to decrease pollutant input to each of the Great Lakes.
Great Lakes Water Quality (GLWQ) Guidance	Promotion of pollution minimization plans to stop pollution before it reaches the environment, and consistent standards to protect human health, wildlife, and aquatic life.
Remedial Action Plans for Great Lakes Areas of Concern (AOCs)	Action-planning process for implementing remedial and preventive actions to restore impaired beneficial uses of specific areas.

pollutants; developing and implementing voluntary and regulatory measures; and developing guidance for evaluating levels of risk of pollutant contamination. These programs have usually involved coordination among various federal, state, and/or local agencies. Though each program is designed to address specific goals, many of these programs coordinate their efforts to ensure that results are not duplicated. For example, the Binational Virtual Elimination Strategy was developed to achieve virtual elimination of persistent toxic pollutants in the Great Lakes, but it also supports and builds upon ongoing processes in the LaMPs, such as the Zero Discharge Demonstration Project through the Lake Superior LaMP. Furthermore, the Great Lakes Emissions Inventories will eventually provide information for determining whether the reduction goals set in the Great Lakes Binational Toxics Strategy have been met. The IADN will monitor whether pollutant levels are actually decreasing.

The Great Lakes programs described in this chapter also complement the Great Waters program in assessing and identifying the extent of atmospheric contamination of hazardous air pollutants to the Great Lakes. Further coordinated progress is needed to improve knowledge and understanding of pollutant contamination, as well as to increase public awareness. In addition to the continuation of the current programs/activities, some high priority efforts for the Great Lakes basin include:

- Improvement in research and monitoring techniques to reduce uncertainties in loading calculations (such as those for the IADN) and therefore, result in better estimates of atmospheric pollutant levels and deposition;

- Improvement in dispersion and deposition models currently being developed to link emission inventory information to atmospheric loadings of Great Lakes pollutants at the water's surface;

- After Lake Michigan Mass Balance data have been analyzed, application of results and modeling tools from the study to the development of a general mass balance model for other hazardous air pollutants;

- Increase in efforts to identify local and long-range sources of Great Lakes pollutants through various source apportionment modeling and emissions inventories, such as in the RAPIDS data base; and

- Continuation of efforts to develop and implement strategies and recommendations to reduce use, generation, and release of pollutants affecting the Great Lakes, particularly through binational efforts such as the reduction challenges proposed under the Binational Virtual Elimination Strategy.

As more information is gathered on the characterization and reduction of atmospheric deposition of toxic pollutants to the Great Lakes, the results of these efforts, as well as the tools used, may be applied to other waterbodies, such as Lake Champlain.

IV.B Lake Champlain

Located around the northernmost borders of the states of New York and Vermont and the southern border of the province of Quebec, Lake Champlain is one of the largest freshwater lakes in the United States, with 1,127 km^2 of surface water, over 70 islands, and 945 km of shoreline (see Figure IV-6). It flows north from Whitehall, New York, almost 193 km across the U.S.-Canadian border to its outlet at the Richelieu River in Quebec, where it joins the St. Lawrence River. Lake Champlain is unique because of its narrow width (19 km at its widest point), great depth (over 122 meters in some parts), and large size of the watershed relative to the lake surface.

The Lake Champlain Basin, composed of the entire watershed or drainage area, spans from the Adirondack Mountains in the west to the Green Mountains in the east, and from the Taconic Mountains in the southeast to the St. Lawrence Valley in the north (Figure IV-6). The total area of the Basin is 21,326 km^2, of which 56 percent is in Vermont, 37 percent is in New York, and 7 percent is in Quebec. The Basin is characterized by an 18:1 ratio of watershed to lake surface area, indicating that the lake represents only about 5 percent of the total basin area. Approximately 89 percent of the Basin is categorized as forest and agricultural land.

By the end of 1994, approximately 645,000 people resided in the Lake Champlain Basin, with the population increasing each year by about 1.2 percent (LCBP 1994). Most of the region is classified as rural, with only Burlington, Vermont, recognized as a metropolitan area (1990 population of 52,000). The Lake Champlain Basin has traditionally had a rural resource-based economy, including agriculture, renewable natural resources (e.g., timber, fish, maple syrup), and non-renewable natural resources (e.g., iron ore, marble, gravel). In recent years, the economy has diversified into other activities, but is still dependent on the natural resources (see sidebar).

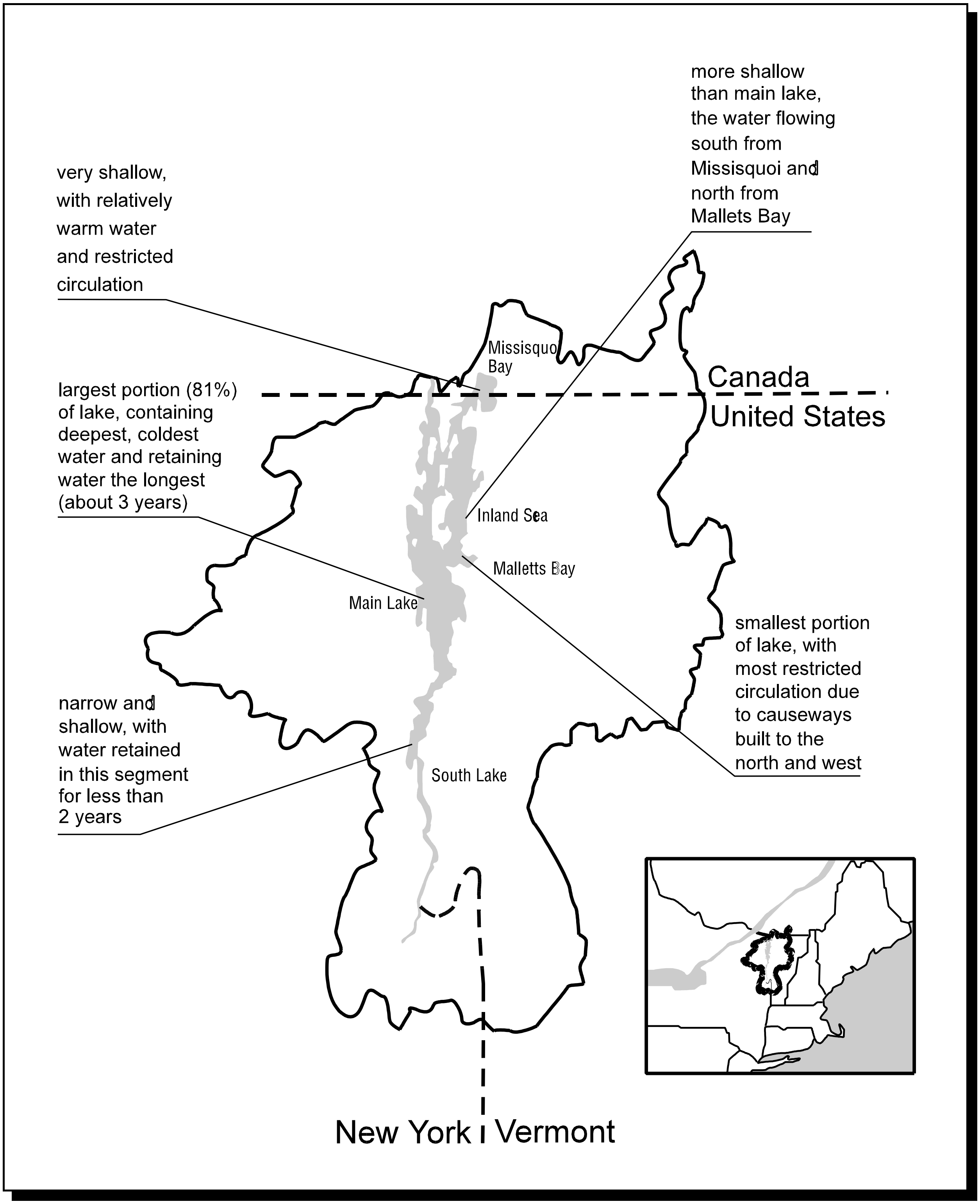
Economic Highlights of Lake Champlain Basin

- Twenty-five percent of the workforce is employed in natural resource-related activities (e.g., agriculture, mining, forestry).
- Resources of the Lake are a major reason why many Basin residents reside in this region.
- Tourism represents a significant economic factor for the region, generating $2.2 billion in 1990, of which 40 percent were Lake related (e.g., marina, white-water rafting). Part of tourism income comes from recreational activities, including $81 million per year from fishing industry (in 1991) and $50 million per year from bird and wildlife viewing (in 1990).

Sources: LCBP 1994, 1996.

Lake Champlain, unlike many other lakes that tend to be more evenly mixed, is made up of five distinct areas or lake segments, each with different physical and chemical characteristics (Figure IV-6). In these lake "divisions," pollutants may accumulate in shallow areas or bays where flushing and water circulation are reduced, and may be deposited at the mouths of rivers where runoff carrying sediment and other pollutants from the watershed is discharged. Human activities also concentrate in many of these areas of the lake, increasing the potential for contamination and exposure.

FIGURE IV-6
Lake Champlain Basin



An understanding of the hydrodynamics of Lake Champlain is essential to predicting how and where pollutants are transported, and where they will end up in the lake. A simplified three-dimensional hydrodynamic transport model, being developed with funding by the Lake Champlain Basin Program (LCBP), will be used as a management tool to determine potential effects of pollutant inputs and other changes to the waterbody (LCBP 1994).

Characterizing Toxic Contaminants in Lake Champlain

Levels of toxic contamination in Lake Champlain are low compared to the Great Lakes; however, concerns for protecting the public health still exist. Fish consumption advisories for two Great Waters pollutants (mercury and PCBs) are currently in effect in both New York and Vermont for fish from Lake Champlain (see Appendix B) (LCBP 1994). Testing of lake bottom sediment near the urbanized sites along the Lake shows pollutants levels that may be of concern and indicates potential risks to aquatic life (see Section II.B). Because of these findings, as well as exceedances of water quality standards set by EPA, NOAA, and the province of Ontario, LCBP gives highest priority to these two pollutants.

The contribution of air deposition as a source of loadings for pollutants, such as metals and organic compounds, is of concern for Lake Champlain and the Basin, and has been the subject of recent studies. Although emissions of toxic pollutants within the Basin are considered low because of the few industries and utilities in the local area, high levels of pollutants may reach the Lake from more distant sources (LCBP 1994, 1996). Data have been limited regarding atmospheric sources, or the movement of pollutants from the atmosphere to the Lake directly or through the watershed.

Atmospheric Sources of Toxic Contamination in Lake Champlain

Local	Mobile emissions Residential energy consumption (e.g., wood burning) Waste incinerators
Regional/ Long-distance	Smelters (Quebec) Utilities (midwestern United States)

A 1990/1991 air monitoring study measured the concentration of toxic metals at four sites in the Lake Champlain Basin (Whiteface Mountain, NY; Willisboro Bay, NY; Burlington, VT; and Underhill, VT) (LCBP 1994). Preliminary data revealed elevated levels of zinc in the air surrounding Burlington, possibly due to refuse incineration, tire wear, and industries. There were also periodic increases in arsenic levels at these and other sites across the Northeast. The source of these arsenic concentrations in the air is believed to be a smelter in Quebec (LCBP 1994). Mercury, lead, and cadmium compounds were also measured in this study; however, there was no indication that the concentrations for these Great Waters pollutants of concern were of concern at the monitoring sites.

The following subsection focuses on currently ongoing mercury research to determine atmospheric deposition to the Lake Champlain Basin. Information on atmospheric deposition of the other high priority pollutant to the basin, PCBs, is lacking at this time.

ATMOSPHERIC DEPOSITION OF MERCURY IN THE LAKE CHAMPLAIN BASIN

Mercury burden in Lake Champlain is evidenced by fish consumption advisories. Direct discharges of mercury are quite limited which has led to concerns that the atmosphere may be the major route of mercury to the lake. Recent studies supported by NOAA and EPA's Great Waters program have investigated the deposition of atmospheric mercury in the Lake Champlain Basin (Burke et al. 1995; Scherbatskoy et al. 1997). The air monitoring data collected from these efforts should provide an initial framework for a more comprehensive analysis of mercury cycling (i.e., deposition, transport, transformation, and accumulation) in the region, and beyond. There is currently no information that compares the estimated amount of mercury entering the lake via water (e.g., runoff) with the amount deposited from the air.

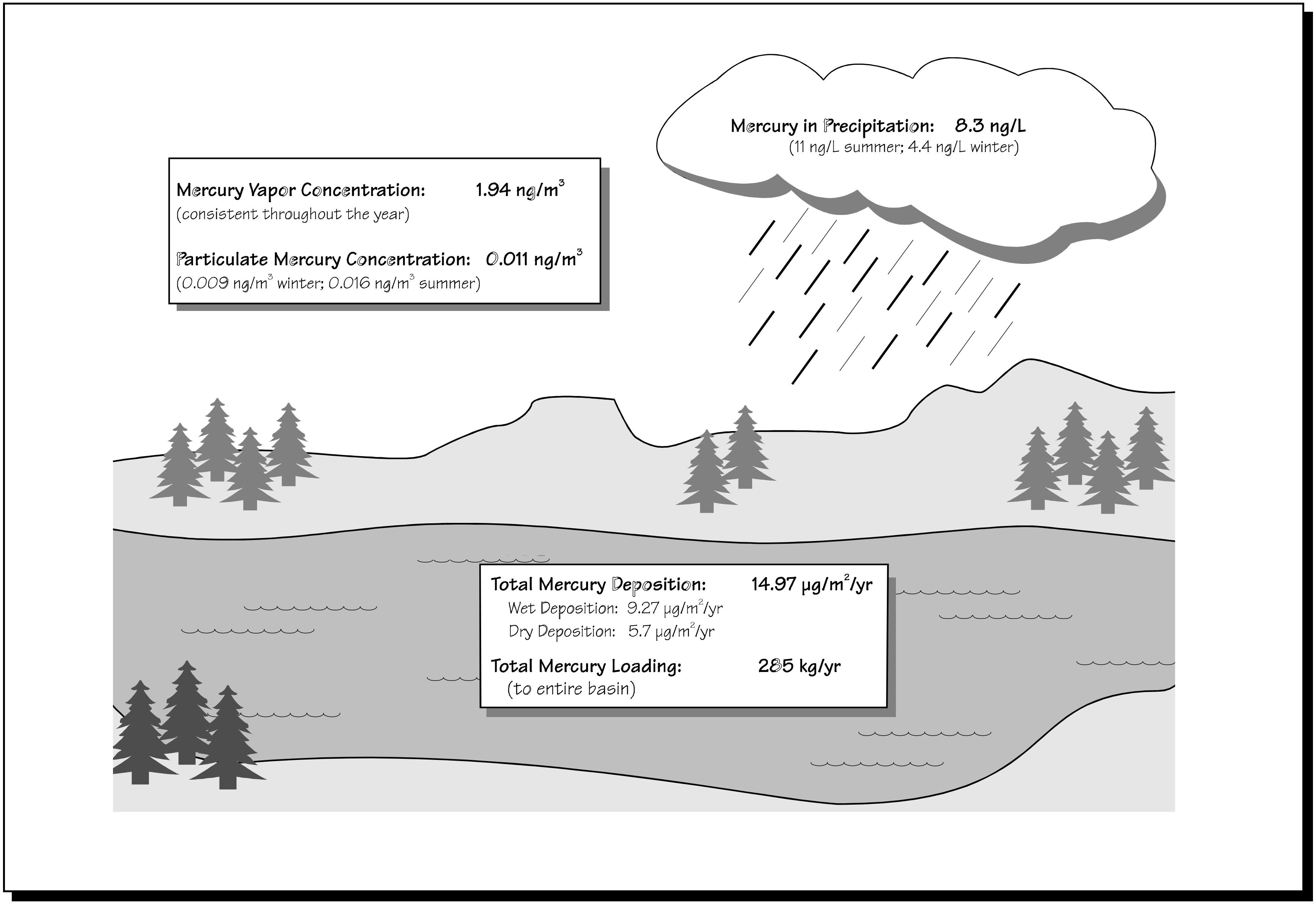
Atmospheric mercury concentration and deposition in Lake Champlain was investigated at a location just east of the Lake in Underhill, Vermont, between 1992 and 1994 (Scherbatskoy et al. 1997). The atmospheric concentration of mercury was measured as gaseous and particulate phases in the ambient air and in precipitation (snow, rain) (see sidebar). Findings on the atmospheric mercury levels near Lake Champlain are presented below and in Figure IV-7:

> **Principal Phases of Atmospheric Mercury**
>
> ***Gaseous (Vapor) Phase Mercury***: consists primarily of elemental mercury (Hg^o; predominant form in the atmosphere), although divalent mercury (Hg^{+2}) may also be present; Hg^{+2} is deposited more rapidly than elemental mercury.
>
> ***Particulate Phase Mercury***: consists of fine particles that are not readily deposited from the atmosphere; bound Hg^{+2} is the predominant form and exists as the nucleus in dust particles.
>
> ***Mercury in Precipitation***: consists of mercury either as dissolved gas or bound to fine particles; primarily particulate Hg^{+2} that has been taken up by rain droplets.

- *Atmospheric gaseous mercury concentration.* Average gaseous concentration for 1993 was 1.94 ng mercury/m^3 (comparable to other sites near the Great Lakes), staying relatively constant throughout the year (Table IV-5). Concentrations are typically two orders of magnitude (100 times) higher than particulate mercury.

- *Average particulate phase mercury concentration.* Unlike the gaseous phase, particulate phase mercury in the air exhibited seasonal variability, with levels higher in winter than in summer (Figure IV-7). The source of the increased particulate mercury concentration in the winter has not been identified, but preliminary meteorological analysis suggests that a more regional influence is important in the transport of particulate phase mercury in the winter, due to higher average wind speeds and colder temperatures during this season. It also is suggested that colder temperatures in the winter may favor condensation of gas onto particles, increasing mercury particulate concentration in the atmosphere (Scherbatskoy et al. 1997). Furthermore, daily mercury concentrations did not fluctuate significantly, as would be expected for strong local sources.

FIGURE IV-7
Atmospheric Mercury in Lake Champlain Basin[a]



[a] Values are annual average, unless otherwise specified.

Source: Scherbatskoy et al. 1997

TABLE IV-5
Comparison of Mean Total Atmospheric Mercury Concentrations [a]
(Gaseous and Particulate Phases and in Precipitation)

Location	Gaseous Phase (ng/m^3)	Particulate Phase (ng/m^3)	Precipitation (ng/L) (volume-weighted)
Lake Champlain	1.94 ± 0.5	0.011 ± 0.007	8.3 ± 5.2
Northern Michigan[b]	NA	0.011	7.9 ± 7.4
Southern Michigan[b]	NA	0.022	10.2 ± 9.8
Northern Wisconsin[c]	1.57 ± 0.4	0.02 ± 0.02	10.5 ± 4.8
Northern Wisconsin[d]	1.8 ± 0.4	0.01 ± 0.02	6.7 ± 5.6

NA = Not Available
[a] Data are means for varying years and periods; different sampling methods for the studies. Gaseous and particulate phase mercury data represent total dry deposition.
[b] Hoyer et al. 1995; Keeler et al. 1995
[c] Fitzgerald et al. 1991
[d] Lamborg et al. 1995

Adapted from Scherbatskoy et al. (1997).

- *Average mercury concentration in precipitation.* Levels appeared to be typically higher in the summer than the winter. Partial data from 1994 suggest that the pattern observed in 1993 is typical for this area. Based on meteorological data, precipitation events with the highest mercury levels during the year were associated with regional transport from the south or west; transport from the west occurred only during the summer months (Burke et al. 1995). The observed mercury levels are low; studies have reported mercury concentration in precipitation generally less than 100 ng/L in areas not directly influenced by emission sources. The type of mercury in precipitation was not quantitatively determined (Scherbatskoy et al. 1997); however, other studies of precipitation found mercury to be in the inorganic (Hg^{+2}) form, probably as mercuric chloride (Burke et al. 1995), with only 2 to 10 percent in the organic (methyl) form (Bloom and Watras 1989; Driscoll et al. 1994).

- *Yearly deposition of mercury.* Deposition to the entire Lake Champlain Basin, not just to surface water, was estimated from precipitation (wet deposition) and dry deposition data. Total annual mercury deposition was not very different from that observed at other U.S. locations (Table IV-6). Dry deposition was calculated by using the same monthly proportion of gas deposition to gas concentration reported in Lindberg et al. (1992). Deposition of particulate phase mercury was not a significant source of atmospheric mercury loadings when compared to deposition of mercury in precipitation and from the gas phase. The atmospheric mercury particulate concentration in the area (0.011 ng/m^3) was much lower than the gas concentration (1.9 ng/m^3); therefore, the estimate of mercury dry deposition was based solely on gaseous mercury (which may be an underestimate of deposition to some extent). As shown in Figure IV-7, the annual dry deposition is 5.7 μg mercury/m^2 (with levels higher in the summer than the winter). Mercury deposition in precipitation for 1993

TABLE IV-6
Comparison of Annual Mercury Deposition Estimates[a]

Location	Estimated Wet Deposition ($\mu g/m^2$/year)	Estimated Dry Deposition ($\mu g/m^2$/year)	Total Mercury Deposition ($\mu g/m^2$/year)
Lake Champlain basin[b]	9.27	5.7	14.97
Michigan[c]	5.5-13.0	NA	NA
Little Rock Lake, WI[d]	4.5 ± 2 from rain 2.3 ± 0.3 from snow	3.5 ± 3	10.3 ± 3.6
Minnesota and Northern Wisconsin[e]	NA	NA	12.5

NA = not available

[a] Methods and assumptions for estimating these values varied with studies.
[b] Scherbatskoy et al. 1997
[c] Hoyer et al. 1995
[d] Fitzgerald et al. 1991
[e] Engstrom et al. 1994; Swain et al. 1992

was 9.27 $\mu g/m^2$, with deposition higher in the summer (1 $\mu g/m^2$/month) compared to winter (0.2 $\mu g/m^2$/month). The pattern for mercury deposition in precipitation is probably a result of higher mercury concentrations and higher amounts of rain during the summer; however, insufficient data are available to determine the source of the increased mercury concentration in the summer.

Atmospheric mercury can also enter the lake from snowmelt, which adds higher concentrations of mercury to the rivers emptying into Lake Champlain. The total mercury concentration at three river sampling sites increased two- to six-fold during the peak snowmelt event as compared to the mean levels over the course of the year (Scherbatskoy et al. 1997).

Because the Lake Champlain basin has such a large ratio of watershed to lake surface area, it is important to understand the deposition and cycling of atmospheric pollutants by the largest categories of land use, forests and agriculture, prior to their entry into the lake in runoff. Unlike the Great Lakes watershed, which has relatively more lake surface area, Lake Champlain's water surface area makes up only five percent of the basin area. Therefore, it is important to understand mercury cycling through the terrestrial, atmospheric, and aquatic systems of the Lake Champlain Basin, as ongoing studies are beginning to address.

SOME PROGRAMS RELATED TO TOXICS EMISSIONS AND REDUCTION IN LAKE CHAMPLAIN

Some programs by states have been established to address toxics reduction and to identify and control source emissions in the surrounding region. These efforts may have important impact on atmospheric deposition to Lake Champlain basin. Some of these programs are highlighted below:

- The State of New York recently began a program for reducing toxics through a multi-media approach. The Multimedia Program for Pollution Prevention, when fully implemented, will integrate environmental protection programs across all "media," such as air, water, and land, to correct the problem with single media programs.

- The State of Vermont conducts a regulatory program for the sources of about 288 hazardous air contaminants (carcinogens, chronic systemic toxicants, and short-term irritants). Hazardous Ambient Air Standards are established for each of these contaminants, with requirements imposed on new and existing sources (excludes fossil fuel combustion) emitting any of these contaminants in excess of a pre-determined "action level" for each pollutant.

- New York and Vermont established a Permit Exchange Agreement in accordance with the 1988 Memorandum of Understanding, in which both states are informed of permitted projects in the basin. It allows the affected public to participate in the comment and review process for the permits. Potential toxics sources subject to this agreement include air pollution sources within 80 km of each state border that annually emit 50 tons of volatile organic compounds, sulfur dioxides, nitrogen oxides, carbon monoxide, or particulate matter, or 5 tons of lead, and/or are subject to Title V of the CAA.

Addressing Toxic Contamination Reduction in Lake Champlain

The Lake Champlain Basin Program (LCBP) (see sidebar) has been the institutional framework for coordinating the development of a comprehensive pollution prevention, control and restoration plan for the future of Lake Champlain. The final plan was released in October 1996 (LCBP 1996), following public meetings that allowed interested parties to comment on the an earlier plan. It is anticipated that the objectives of the final plan will be implemented by the Lake Champlain Steering Committee, which is represented by environmental officials from New York, Vermont, and Quebec.

> **Lake Champlain Basin Program**
>
> The Lake Champlain Special Designation Act, sponsored by senators from Vermont and New York, was signed in 1990 and states that Lake Champlain is a resource of national significance. The intent of the Act is to create a comprehensive plan for protecting the future of Lake Champlain and its watershed. The coordination of the activities stated in the Act is the responsibility of the Lake Champlain Basin Program (LCBP), which is jointly administered by the U.S. EPA, the States of Vermont and New York, and the New England Interstate Water Pollution Control Commission. EPA was given $10 million in funds for five years to develop a comprehensive pollution prevention, control, and restoration plan for Lake Champlain; the final plan was released in 1996. Other cooperating agencies include the U.S. Fish and Wildlife Service, U.S. Department of Agriculture, U.S. Geological Survey, NOAA, and National Park Service. Formal involvement of Quebec is through the Lake Champlain Steering Committee.

One major issue addressed in the plan is the prevention of pollution from toxic substances in order to protect public health and the Lake Champlain ecosystem. Implementation of the plan will require coordination along all levels of government, organizations, and individuals. The plan identified the following major technical and policy issues involved in determining the most appropriate and cost-effective actions to reduce toxic contamination of Lake Champlain.

♦ *Define scope of toxics reduction effort.* Research is needed to define the extent of problems related to toxic pollution in tributaries and in the air. Efforts should be made to improve source identification, with attention given to reducing both nonpoint and point sources through all media and remediating current sources of contamination throughout the Basin.

♦ *Focus efforts on Lake Champlain pollutants of concern and sites of concern.* The List of Toxic Substances of Concern was established by LCBP, which identified mercury and PCBs as Group 1 chemicals and meriting highest priority for management action (see sidebar). These two pollutants are found in the sediment, water, and biota at levels above appropriate standards or guidelines in Lake Champlain. Because of limited resources to study and monitor toxic substances, assessments should focus on specific sites where contamination is known.

Toxic Substances of Concern in Lake Champlain by Priority Group

Group 1: PCBs, mercury

Group 2: Arsenic, cadmium, chromium, dioxins/furans, lead, nickel, PAHs, silver, zinc

Group 3: Ammonia, persistent chlorinated pesticides, phthalates, chlorinated phenols, chlorine, copper

Group 4: Other contaminants known to be used or known to occur in the Basin (e.g., volatile organic compounds such as benzene, pesticides such as atrazine, strong acids and bases)

Source: LCBP 1996.

♦ *Identify sources and quantify loads of toxic substances.* Efforts to determine sources of toxic substances within Lake Champlain have been initiated, and few "active" sources have been identified. Two major information gaps in this area that need to be addressed include the extent of contamination from outside the basin, and the role of historical sources (e.g., discharged lead batteries released into the waterbody) and contaminated sediment.

♦ *Adopt strategy to prevent pollution.* Pollution prevention techniques (e.g., source reduction) may be used to achieve reductions at the source of the problem, and eventually reduce pollutants in the lake. Vermont and New York have initiated programs to accomplish this goal, although these programs are not yet integrated into the existing pollution control programs.

♦ *Establish firm and defensible toxic reduction goals.* The current chemical-by-chemical approach to managing pollution in Lake Champlain cannot account for the impact of cumulative or combined effects, and does not protect against unregulated (and potentially more toxic) chemicals entering the lake; therefore, the reduction strategy should be expanded to address toxic substances that do not yet exceed human health standards or cause measurable impacts within the basin.

In addition, as part of these efforts, relevant information gathered from other programs, such as those initiated in the Great Lakes, should be applied to Lake Champlain. As discussed in Section IV.A, the Great Lakes Water Quality Agreement Parties adopted a long-term goal for virtual elimination of sources of specific pollutants. This effort demonstrates the importance of binational cooperation to address concerns related to reducing toxic emissions in the Great Lakes, as well as in Lake Champlain.

IV.C Chesapeake Bay

The Chesapeake Bay, largest of the 130 estuaries in the United States, was the first in the nation to be targeted for restoration as an integrated watershed, airshed, and ecosystem. The 166,000 km^2 drainage basin (or watershed), shown in Figure IV-8, covers parts of six states (Delaware, Maryland, New York, Pennsylvania, Virginia, and West Virginia) and the District of Columbia, and includes more than 150 rivers and streams. The major tributary basins within the Chesapeake Bay watershed are shown in Figure IV-9, which is found later in this section.

Stretching from Havre de Grace, Maryland, to Norfolk, Virginia, the Chesapeake Bay is 314 km long, and ranges from 5 to 56 km wide. The Bay has over 90,000 km of shoreline (more than the entire West Coast of the continental United States) and a surface area of approximately 30,800 km^2. Generally shaped like a shallow tray, the Bay's average depth, including all tidal tributaries, is only 6 meters, with a few deep troughs running along much of its length that average 18 to 21 meters, and reaching 53 meters at the deepest point. To visualize the relatively large watershed in contrast to the small Bay volume, imagine that the Bay's watershed is reduced to the size of this page; the relative size of the Chesapeake Bay would be a section 7.2 inches by 0.9 inches in the lower right hand corner, and the average depth of the Bay would be represented by one sixtieth the thickness of the paper (see also Figure IV-8).

Economic Highlights of Chesapeake Bay

- In 1992, the dockside value of commercial shellfish and finfish harvests from Chesapeake Bay was close to $80 million.
- In 1993, more than 175,000 pleasure craft (e.g., sail boats) were registered in the Bay.
- Close to 1 million anglers in Maryland and Virginia made an estimated 600,000 fishing trips in 1991. Recreational fishing in these states is estimated at more than $1 billion annually.
- The Chesapeake is a key commercial waterway, and home to two of the nation's five major North Atlantic ports (Port of Baltimore, MD, and Hampton Roads Complex, VA). More than 90 million tons of cargo were shipped via the Bay in 1992.

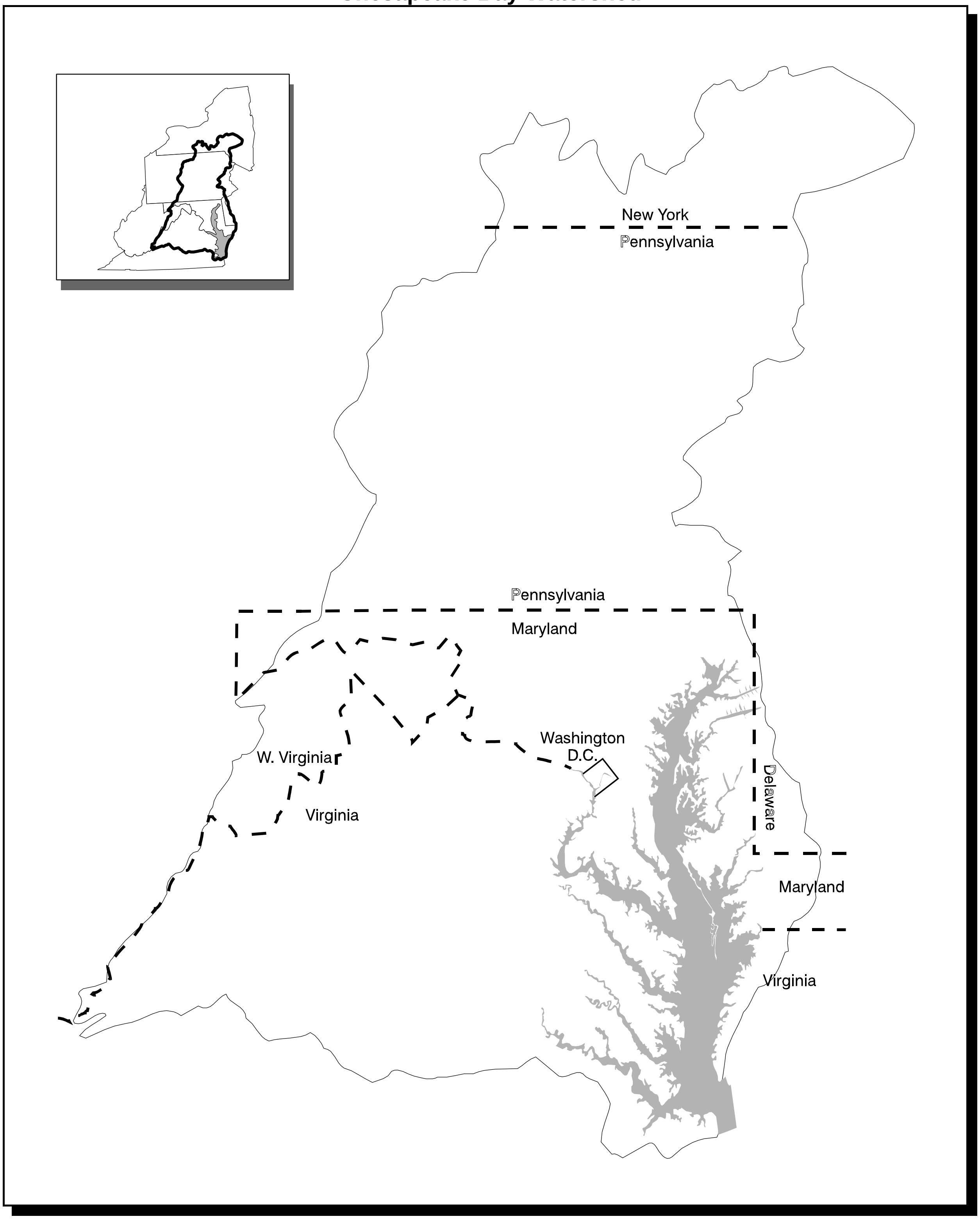
Supporting 295 species of finfish, 45 species of shellfish, and 27,000 plant species, the Chesapeake Bay is a national ecological treasure. The Chesapeake Bay is also home to 29 species of waterfowl and is a major resting ground along the Atlantic Migratory Bird Flyway. Every year, one million waterfowl winter in the Bay's basin. Economic highlights of the Chesapeake Bay are presented in the sidebar above. In all, the Chesapeake is a commercial and recreational resource for more than 14 million Bay basin residents.

The remainder of Section IV.C presents information on: the Chesapeake Bay Program; atmospheric deposition of nitrogen to the Chesapeake Bay; and atmospheric deposition of toxic contaminants to the Chesapeake Bay.

Chesapeake Bay Program

Now in its fourteenth year, the Chesapeake Bay Program is a unique, regional partnership that has directed and coordinated Chesapeake Bay restoration since the signing of the historic 1983 Chesapeake Bay Agreement. The principal partners in the Chesapeake Bay Program include the State of Maryland, the Commonwealths of Virginia and Pennsylvania,

FIGURE IV-8
Chesapeake Bay Watershed



the District of Columbia, the Chesapeake Bay Commission (representing the state legislatures), and EPA on behalf of the federal government.

In 1983, EPA identified an excess of the nutrients nitrogen and phosphorus, and the resulting accelerated eutrophication, as the primary reason for the decline in water quality in Chesapeake Bay (U.S. EPA 1983). Excess nutrients stimulate "blooms" of phytoplankton algae, which then sink to the bottom of the Chesapeake. In the bottom waters, decay of the phytoplankton consumes oxygen, which expands the area of anoxic bottom waters (i.e., "dead waters" largely devoid of oxygen and unable to support life). Blooms of algae also reduce light to submerged aquatic vegetation (SAV), resulting in the loss of an important habitat for juvenile fish and crabs. (A recent assessment of the state of the Bay is presented in the sidebar.)

The state of the Chesapeake restoration and protection effort was described in the latest *State of the Chesapeake Bay* report (CBP 1995b):

"If the health of the Bay could be likened to that of a hospital patient, the doctor would report that the patient's vital signs, such as living resources, habitat, and water quality, are stabilized and the patient is out of intensive care. Some vital signs, such as striped bass and Bay grasses have improved dramatically, while a few, such as oysters, are in decline. Other vital signs are mixed but stable. Nutrients are being reduced, with phosphorus levels down considerably more than nitrogen levels and dissolved oxygen remains steady. Overall, the patient still suffers stress from an expanding population and changing land use, but it is on the road to recovery. Taken as a whole, the concentrated restoration and management effort begun ten years ago has produced tangible results--a state of the Bay that is better today than when we started..."

Using the watershed as the central focus, the Chesapeake Bay Agreement of 1983 recognized the historical decline of the Bay's living resources and recommended a cooperative approach among the federal and state governments within the watershed to address problems defined by the 1978-1983 Chesapeake Bay Research Program. The one-page agreement committed the signatories to work together to "fully address the extent, complexity, and sources of pollutants entering the Bay." The watershed approach of the state-federal partnership was chosen as the most practical method for implementing restoration efforts on both a local and regional scale.

Building on an expanded understanding of the Bay system and increasing experience with on-the-ground implementation within the cooperative basinwide partnership, a new Chesapeake Bay Agreement was signed in 1987 that set forth a comprehensive array of goals, objectives, and commitments to address living resources, water quality, growth, public information, and governance (Chesapeake Executive Council 1987). The centerpiece of the agreement was a commitment to achieve a 40 percent reduction of nitrogen and phosphorus entering the Bay by the year 2000. This measurable goal added a specific direction to ongoing monitoring, modeling, and nutrient reduction implementation programs. Through the 1987 Bay Agreement, the signatories also committed to "quantify the impacts and identify the sources of atmospheric inputs on the Bay system." This seemingly minor commitment at the time set the stage for a decade-long path to formally address atmospheric deposition as an integral component of basinwide pollution reduction strategies and implementation actions.

Atmospheric Deposition of Nitrogen to Chesapeake Bay

This section presents information on the Chesapeake Bay Nutrient Reduction Strategy, an overview of atmospheric nitrogen loadings to the Bay (from modeling of the airshed, to nitrogen loadings estimates, to modeling of the watershed and estuary), and areas of uncertainty and work underway. Although the Chesapeake Bay Agreement and the baywide Nutrient Reduction Strategy focus on two main nutrients, nitrogen and phosphorus, this section focuses mainly on nitrogen because atmospheric deposition, the focus of this report, is a significant pathway of concern for nitrogen loadings only.

NUTRIENT REDUCTION STRATEGY

The Chesapeake Bay Agreement commits the signatories to reduce the "controllable" nutrient loads by 40 percent by the year 2000. Controllable loads are defined as the baseline year loads minus the loads delivered to the Bay under an all- forested watershed (i.e., a watershed providing only natural, uncontrollable sources of nitrogen) within the Bay Agreement signatory jurisdictions (Linker et al. 1996). In other words, controllable loads are defined as everything over and above the total phosphorus or total nitrogen loads that would have come from an entirely forested watershed in the States of Pennsylvania, Maryland, and Virginia, and the District of Columbia, given existing rates of atmospheric deposition. In this definition, point source loads are considered entirely controllable. In addition, for the Bay Agreement, emissions of nitrogen compounds leading to atmospheric deposition are considered uncontrollable. Nonpoint sources may be controllable or uncontrollable.

Sources of Nitrogen Entering the Bay

Sources of the 170.8 million kilograms of nitrogen delivered annually to the Bay include:

- Point source water discharges (23% or 39.3 million kg), such as sewage treatment plants;
- Atmospheric deposition directly to tidal waters(9% or 15.4 million kg) and indirectly to tidal waters (18% or 30.2 million kg); and
- Other nonpoint sources (50% or 85.9 million kg), such as runoff from agriculture and urban areas.

To measure the goal of reducing controllable nutrient loads by 40 percent, the Chesapeake Bay Program established a 1985 baseline of nutrient loads. The 1985 baseline load was defined using 1985 point source loads and a 1984-1987 average load for nonpoint sources. The Chesapeake Bay Program chose the average load of the 1984-1987 period as the base to be representative of nonpoint source loads for all tributaries, because river flow and associated nonpoint source loads may vary depending on rainfall. Table IV-7 presents the 1985 base load and 40 percent reduction target for the major tributary basins of the Bay, and Figure IV-9 presents the locations of the tributary basins. After the year 2000, the tributary nutrient reduction targets (i.e., the 1985 base load minus the 40 percent reduction target) become nutrient caps that are not to be exceeded at any time in the future even in the face of continued population growth and development of the watershed.

In 1992, the basinwide reduction goal was reevaluated and allocated among the ten major tributary watershed basins. The state jurisdictions, with direct involvement of the public, then developed comprehensive tributary-specific nutrient reduction strategies within the individual watersheds. As part of the 1992 amendments to the Chesapeake Bay Agreement, the signatories committed "to incorporate in the Nutrient Reduction Strategies an air deposition component which builds upon the 1990 Amendments to the federal Clean Air Act and explores additional implementation opportunities to further reduce airborne sources of nitrogen entering Chesapeake Bay and its tributaries" (Chesapeake Executive Council 1992).

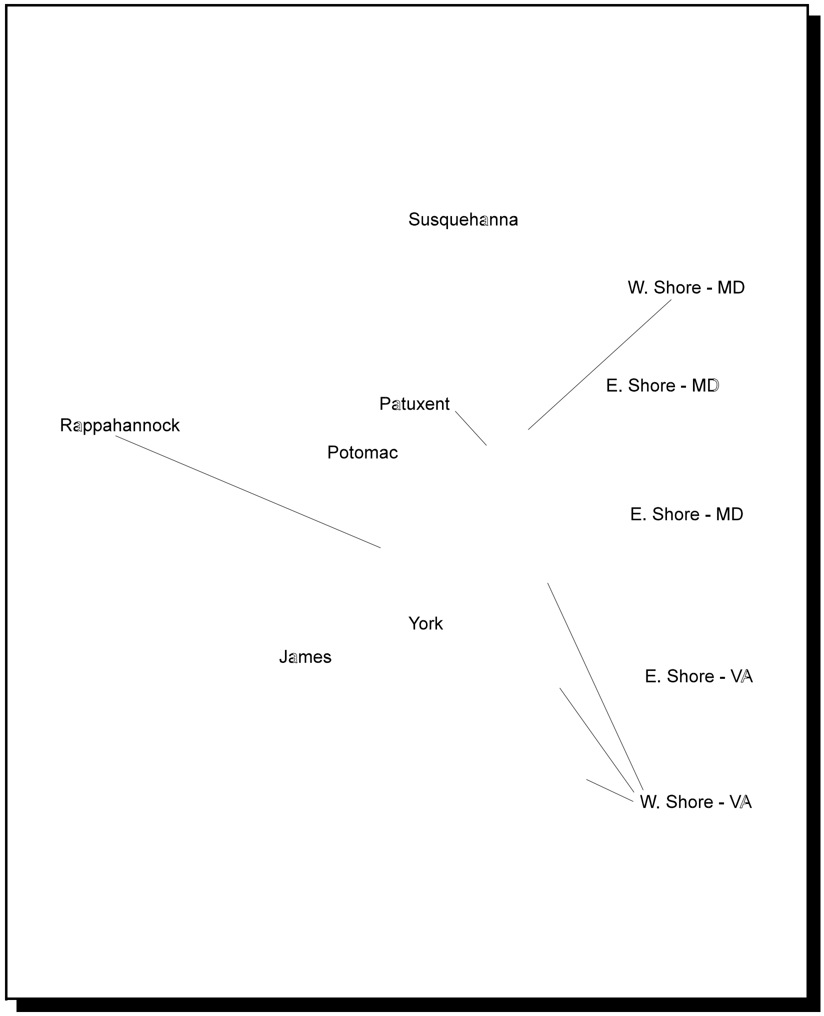
TABLE IV-7
Chesapeake Bay Basin Nutrient Reduction and Loading Caps by Major Tributary Basin
(in millions of kilograms)

Major Tributary Watershed Basin	Nutrient	1985 Base Load	40% Target Reduction	Year 2000 Agreement Loading Cap
Eastern Shore MD	Nitrogen	10.34	2.54	7.80
	Phosphorus	0.82	0.28	0.54
Eastern Shore VA	Nitrogen	0.82	0.18	0.64
	Phosphorus	0.04	0.01	0.03
James[a]	Nitrogen	19.82	6.39	13.43
	Phosphorus	2.80	0.97	1.83
Patuxent	Nitrogen	2.22	0.64	1.59
	Phosphorus	0.24	0.09	0.15
Potomac[b]	Nitrogen	31.16	8.48	22.68
	Phosphorus	2.41	0.78	1.64
Rappahannock	Nitrogen	3.76	1.18	2.59
	Phosphorus	0.39	0.15	0.24
Susquehanna[c]	Nitrogen	52.98	8.30	44.68
	Phosphorus	2.69	1.01	1.69
York	Nitrogen	2.90	0.86	2.04
	Phosphorus	0.42	0.15	0.27
Western Shore MD	Nitrogen	12.02	4.39	7.62
	Phosphorus	0.77	0.30	0.47
Western Shore VA	Nitrogen	1.91	0.54	1.36
	Phosphorus	0.23	0.09	0.14

[a] James loads include only loads from Virginia.
[b] Potomac loads include only loads from Pennsylvania, Maryland, Virginia, and the District of Columbia.
[c] Susquehanna loads include only loads from Pennsylvania and Maryland.

Source: Adapted from CBP 1992.

FIGURE IV-9
Major Tributary Basins of the Chesapeake Bay

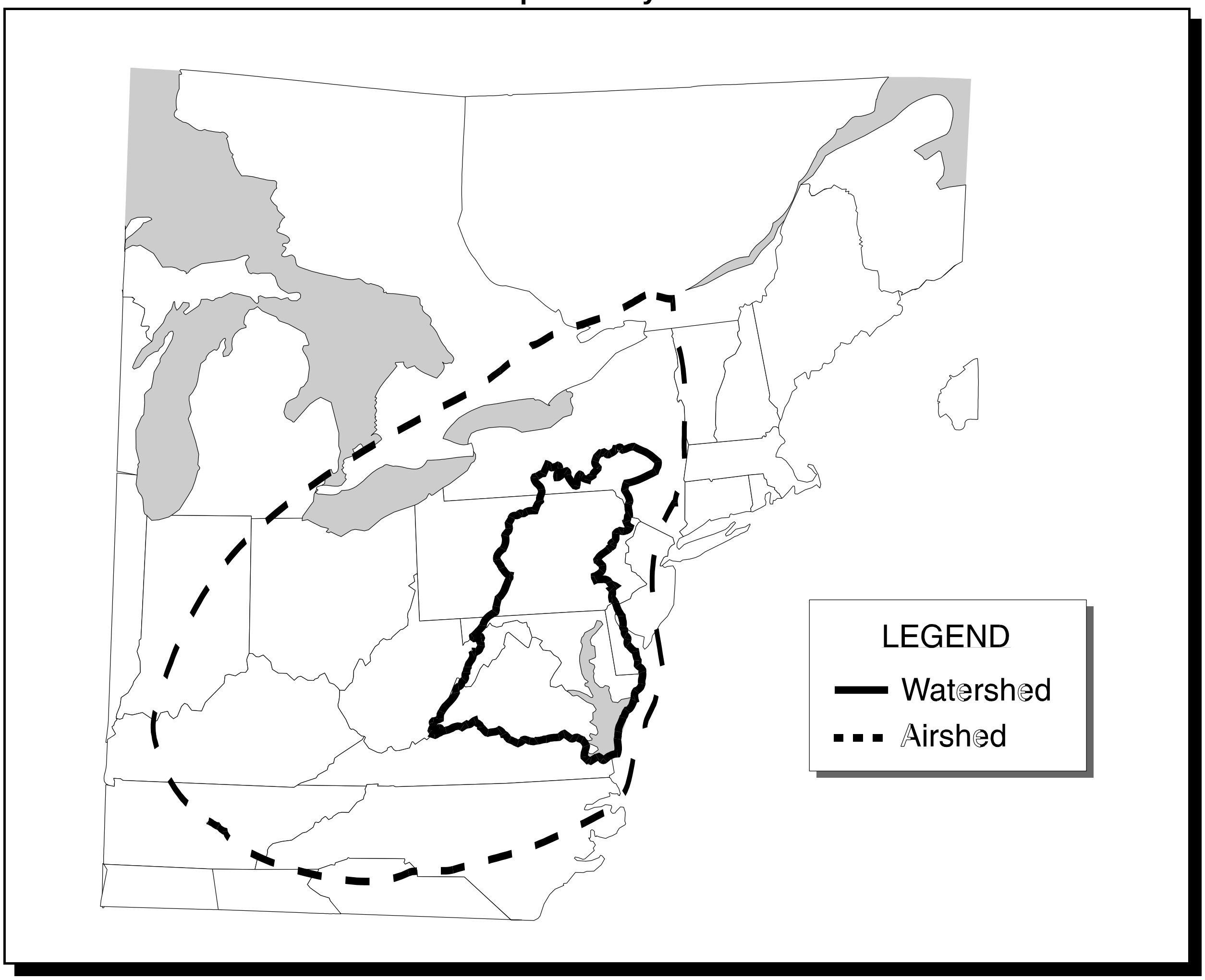


Any reductions in nitrogen loads brought about by programs implementing the CAA are considered to be additional nutrient load reductions separate from the point and nonpoint source reductions identified in the tributary nutrient reduction strategies. CAA implementation is expected to reduce nitrogen loads in Chesapeake Bay beyond the tributary strategy reductions (CBP 1994a). However, these additional reductions may last only a short time; at some time after the year 2000, population growth and increased land development are expected to begin eroding the gains made by the CAA. This expected increase in nutrient loads may make it difficult to meet the caps on nutrient loads to the Bay. Indeed, it was acknowledged at the time the tributary strategies were developed that "achieving a 40 percent nutrient reduction goal, in at least some cases, challenges the limits of current point and nonpoint source control technologies" (Chesapeake Executive Council 1992). To maintain the restoration progress that will be achieved by the year 2000, the technological limits of controls on reductions from point and nonpoint sources may have to be expanded to make further reductions in these areas economically attractive, or other sources of controllable nutrients may have to be considered to achieve cost-effective ecosystem protection in the Bay.

MODELING AIR TO WATERSHED TRANSPORT: THE CHESAPEAKE BAY AIRSHED

A series of linked computer models have been developed by the Chesapeake Bay Program to simulate the transport of nitrogen from its emission sources to the Chesapeake Bay watershed and eventually into the tidal Bay waters. As a first step in establishing the air to tidal waters connection, the "airshed" of the Chesapeake Bay was defined. The boundaries of the airshed were defined as the contiguous areas whose sources "significantly" contributed (i.e., 75 percent) to atmospheric deposition of nitrogen to the Bay and its surrounding watershed (Dennis 1997). These boundaries were delineated by running a series of scenarios on the Regional Acid Deposition Model (RADM), using a predefined point of diminishing return (i.e., when a 50 percent reduction in emissions from large source regions would be expected to produce less than a 10 percent reduction in deposition onto the Bay watershed). The resulting 906,000 km^2 airshed, shown in Figure IV-10, is about 5.5 times larger than the Bay's watershed and includes: all of Maryland, Virginia, Pennsylvania, Delaware, the District of Columbia, West Virginia, and Ohio; most of New York; half of New Jersey, North Carolina, and Kentucky; and parts of Tennessee, South Carolina, Michigan, Ontario, and Quebec (including Lakes Erie and Ontario). (See Chapter III for a description of RADM and Dennis (1997) for more information on the use and limitations of RADM in this study.)

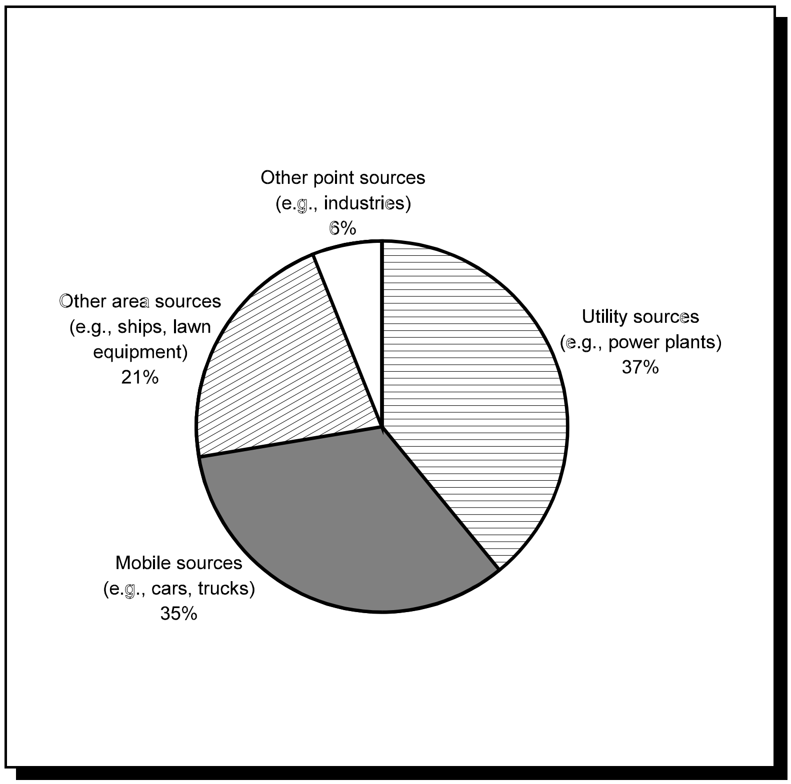
FIGURE IV-10
Chesapeake Bay Airshed



According to the Bay airshed model, about 25 percent of the nitrogen that is deposited on the Bay and its surrounding watershed originates from sources within the Bay watershed. Sources located within the jurisdictions of the Bay Agreement signatories of Maryland, Virginia, Pennsylvania, and the District of Columbia (including those sources that are within the state boundaries but outside of the Bay watershed) contribute about 40 percent of the nitrogen that deposits on the Bay and its watershed (Dennis 1997).

As defined, the Bay airshed, which accounts for 30 percent of all nitrogen emissions in the eastern United States and Canada, accounts for 75 percent of the atmospheric nitrogen deposited onto the Bay and its watershed. The remaining 25 percent of the deposition originates from emission sources outside the defined airshed (Dennis 1997). Therefore, the Chesapeake Bay airshed as defined here is smaller than the actual areas of the United States and Canada that contribute to nitrogen deposition to the Bay watershed. A still unresolved portion of the airshed is the portion that contributes to atmospheric deposition to offshore ocean waters which, in turn, contributes to the influx of nitrogen from coastal waters into the southern Chesapeake Bay (CBP 1994b).

FIGURE IV-11
NO_x Emission Sources in the Major Bay Influencing States



Researchers compared results from the Bay airshed model to emissions inventory data on sources of NO_x emissions and evaluated the contribution of these sources to nitrogen loads to the Bay. As shown in Figure IV-11, data from the emissions inventory indicate that the contributions from utility and mobile sources in the major Bay influencing states (i.e., Maryland, New Jersey, New York, Ohio, Pennsylvania, Virginia, West Virginia) to NO_x emissions are roughly equal and make up the majority of emission sources. Through RADM, these data were confirmed and the patterns of nitrate deposition from the two sources were simulated. The model simulations suggest that utilities contribute a majority of the nitrate that deposits on the western side of the Bay watershed and that nitrate deposition from utility emissions shows a decreasing trend from the western to eastern portion of the watershed (see Figure IV-12[11]). These simulations further suggest that mobile sources, associated with NO_x emissions from the Boston to Washington, D.C., metropolitan areas, contribute a majority of the nitrate that deposits along the Delmarva Peninsula, the Chesapeake Bay itself, and the lower portions of the western shore tidal tributaries (see Figure IV-13). In contrast to utility sources, the simulated deposition from mobile sources shows a decreasing trend from the eastern to western portion of the basin. Model scenarios simulating the effects of a uniform 50 percent reduction in nitrogen emissions from utilities alone and then from mobile sources alone show the same west to east, or east to west, gradients respectively (Dennis 1997).

[11] In Figures IV-12 and IV-13, a rough outline of the watershed and airshed is also shown. Each shaded area in these figures represents the percentage of all emissions that emissions from sources within the shaded area contribute to nitrogen oxides that deposit to the Bay.

FIGURE IV-12
RADM Total (Wet and Dry) Nitrate Deposition from Utility Sources
(as a percent contribution of 1990 Base Case)

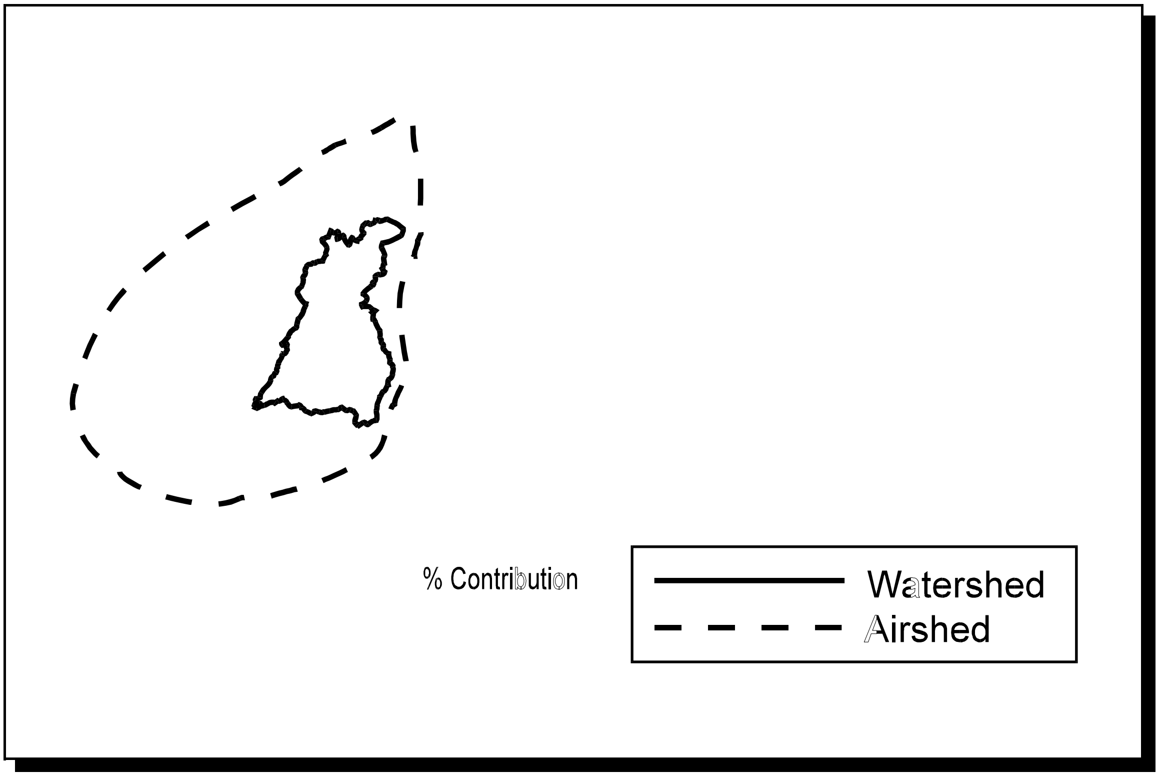
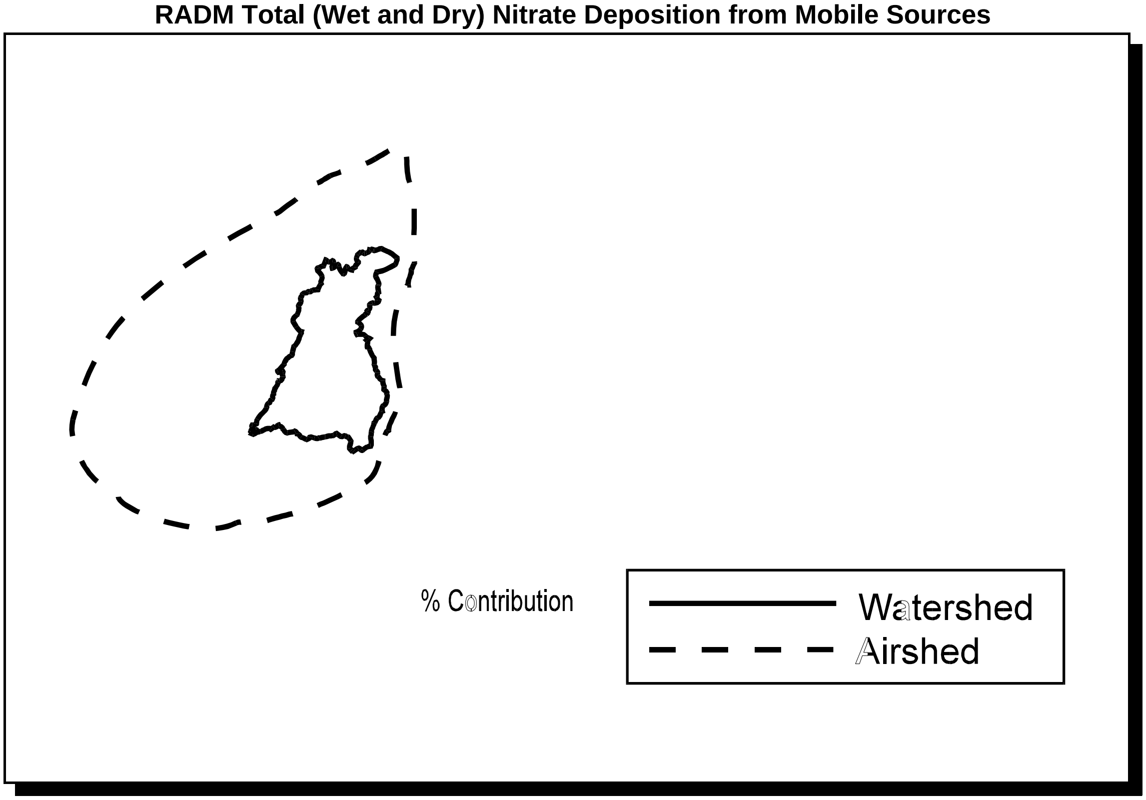


FIGURE IV-13
RADM Total (Wet and Dry) Nitrate Deposition from Mobile Sources



ATMOSPHERIC NITROGEN LOADINGS TO CHESAPEAKE BAY

Atmospheric nitrogen loads from the airshed are transported to the Chesapeake Bay by three routes: direct deposition, both wet and dry, to the Bay tidal waters (i.e., direct loadings); indirect deposition, both wet and dry, to the watershed with subsequent runoff and river transport to the Bay (i.e,. indirect loadings); and deposition, both wet and dry, to adjacent offshore coastal waters with subsequent transport to the Bay through coastal currents. The first two processes, direct and indirect deposition to the Bay, are discussed below, as are some estimates of total loadings to the Bay using both a mass balance approach and computer models. The third pathway, deposition to offshore coastal waters, is the least understood route and is discussed later in this section under areas of uncertainty. Different nitrogen compounds that are measured or estimated in nitrogen loadings are discussed in the box on the next page.

Direct Loadings. The first estimates of atmospheric deposition to the tidal waters of Chesapeake Bay were made through spatial extrapolation of the National Atmospheric Deposition Program (NADP) sites in the Chesapeake watershed (Cerco and Cole 1994). The NADP is a long-term nationwide monitoring program that was started in the 1970s. Based on the annual loads reported by NADP, and an assumption that dry deposition of nitrate is equal to the long-term average of wet deposition of nitrate (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988), atmospheric deposition of inorganic nitrogen to the tidal waters of the Chesapeake was estimated as 6.4 million kilograms of nitrate and 1.8 million kilograms of ammonia. In addition, organic nitrogen was estimated as 6.8 million kilograms (Smullen et al. 1982). Studies have explored the idea that atmospheric deposition may contribute a significant proportion of phytoplankton nitrogen demands in coastal areas (Paerl 1985; Paerl 1988; Paerl et al. 1990); phytoplankton require nitrogen, both new and recycled, for growth. Fogel and Paerl (1991), for example, have estimated that 20 to 50 percent of annual new nitrogen demands for phytoplankton in Albemarle-Pamlico Sound, NC, may be supplied by direct atmospheric deposition to the water surface (wet and dry).

Using NADP data, wet deposition directly to the Chesapeake Bay tidal surface waters has been estimated to range from 3.3 to 4.2 million kilograms of nitrate per year (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988). Though NADP monitoring data allow initial estimates to be made of atmospheric deposition to the tidal Bay, it is not currently known if the over-land measurements of wet deposition accurately represent over-water wet deposition. To investigate this question, a daily precipitation chemistry site was established on Smith Island, Maryland, in late 1995. This site is providing the first time-series measurements of over-water wet deposition collected on the east coast.

Although the dry deposition to surface water loading rates of nitrogen compounds have been estimated for most nitrogen species over open ocean (Galloway 1985; Duce et al. 1991), these rates may not apply to coastal areas because of the different meteorological processes involved. Through the use of instrumented Chesapeake Bay Observing System (CBOS) buoys owned by the University of Maryland, estimates of nitrogen (HNO_3, NO_2, NH_4) dry deposition rates to the Bay tidal surface waters have been developed (Valigura 1995). These estimates corroborate those given by other investigators to some extent, but still cover a wide range of values, from 0.7 to 2.2 million kilograms of nitrate per year. From this data set, calculations were performed to determine the effect of atmospheric dry deposition on phytoplankton dynamics. This analysis demonstrated that dry deposited nitrogen may provide 10 percent of the annual "new nitrogen" demands by phytoplankton in Chesapeake Bay, and that individual events could supply up to 75 percent of the new demands for periods of several days (Malone 1992; Owens et al. 1992).

Characterization of Nitrogen Compounds

Most atmospheric nitrogen compounds (excluding N_2 and N_2O, which are relatively unreactive in the lower atmosphere) fall into two categories: reactive nitrogen and reduced nitrogen. In addition, some organic nitrogen species arise in the atmosphere from the interaction between nitrogen oxides and certain hydrocarbons. The relative portions of the different nitrogen forms in nitrogen loadings can vary widely based on source types and locations, proximity of sources to receiving waters, atmospheric transport, and physical and chemical transformations. Current estimates are that reactive nitrogen is the largest contributor to atmospheric deposition in coastal waters (40 to 60 percent), with ammonia (20 to 40 percent) and organic nitrogen (0 to 20 percent) also contributing significant amounts.

Reactive nitrogen compounds, primarily oxides of nitrogen, are emitted to the atmosphere through both natural and anthropogenic pathways, overwhelmingly (95 percent) as nitric oxide (NO). Natural NO sources include emissions from soils and generation by lightning; dominant anthropogenic sources include emissions from automobiles, power plants, and biomass burning. The dominant source of reactive nitrogen oxides present in air over North America is high-temperature combustion (e.g., power plants, automobiles). NO generated by combustion reacts quickly in the lower atmosphere, forming nitrogen dioxide (NO_2). The NO_2 is rapidly converted back to NO by ultraviolet light (photochemistry), then NO reacts again, resulting in a cycle driven by volatile organic compounds. From this photochemical cycle, ozone (O_3) is produced. The cycle is broken when NO_2 terminates into stable products, principally nitric acid vapor (HNO_3), and the NO gets used up. The ozone issue is therefore intimately related to the NO_x (defined as NO + NO_2) question. NO_2 slowly deposits to the underlying surface (too slowly to break the cycle), but nitric acid vapor (HNO_3) is easily and quickly deposited. HNO_3 is the source of most of the reactive nitrogen deposited to the earth's surface.

Reduced nitrogen compounds include ammonia (NH_3) and ammonium (NH_4^+). NH_3 is emitted into the atmosphere through both natural and anthropogenic pathways. Natural sources of NH_3 include microbial decomposition of organic nitrogen compounds in soils and ocean waters and volatilization from animal and human wastes. Anthropogenic sources include the manufacture and release of commercial and organic fertilizers during and after application and fossil fuel combustion. Human activities such as manure management and biomass burning exacerbate emissions from otherwise natural processes. NH_3 is a highly reactive compound and has a short residence time in the atmosphere. It is primarily emitted at ground level and quickly deposits to the area near its source unless it reacts with other gaseous chemicals (e.g., SO_2, HNO_3) and is converted to NH_4^+ aerosol (Asman 1994; Langland 1992). NH_4^+ can be transferred regionally as ammonium salts, such as ammonium nitrate NH_4NO_3 and ammonium sulfate $(NH_4)_2SO_4$, and these salts are the primary contributor to NH_4^+ concentrations measured in precipitation. Scavenging of NH_3 by precipitation can also be a major source of NH_4^+ in precipitation.

Organic nitrogen may be a significant fraction of the total nitrogen measured in precipitation (Cornell et al. 1995; Gorzelska et al. 1997; Milne and Zika 1993). Data on the deposition of organic nitrogen has been limited, however, because of the paucity of reliable measurements, the historical variability in analytical techniques and results, and the current lack of suitable and uniform analytical measurement techniques. In fact, only wet deposition of dissolved organic nitrogen (DON) has been addressed. Various estimates for the relative flux of organic versus total nitrogen via wet deposition range from less than 10 percent to greater than 60 percent. The contribution of the unresolved organic fraction may significantly augment the atmospheric deposition of nitrogen to coastal waters. However, in addition to the lack of dry deposition data, there remain many conceptual questions related to source identification and the bioavailability of atmospheric organic nitrogen.

Sources: Luke and Valigura 1997; Paerl et al. 1997; Valigura et al. 1996.

Indirect Loadings. Quantifying indirect loadings of nitrogen to Chesapeake Bay, which refers to the atmospheric deposition of nitrogen to the terrestrial watershed and subsequent transport of the nitrogen from the terrestrial watershed to Bay surface waters, is an important component of the estimate of total atmospheric deposition of nitrogen to the Bay, yet it is largely uncertain. NADP monitoring data provide an initial estimate of the atmospheric deposition to the Chesapeake Bay watershed. Generally, higher deposition levels are found in the northern portions of the basin. In fact, some of the highest readings for atmospheric deposition of nitrate in the NADP monitoring network come from the northern sections of the Chesapeake basin. Greatest uncertainty is in dry deposition of nitrogen, which is not routinely measured by NADP.

The amount of atmospheric nitrogen transferred to surface waters within a given watershed depends on land use, total nitrogen loading from atmospheric, fertilizer, animal waste, and biosolid sources, the amount of soil nitrogen, characteristics of the soil, site rainfall and temperature, the elevation and slope of the land, and the type, age, and health of the vegetative cover. These characteristics vary independently, making it difficult to determine the fate of atmospherically deposited nitrogen over any area of significant size. However, several classification schemes for forested sites have been developed to evaluate a site's potential to retain and leach nitrogen (Melillo et al. 1989; Johnson and Lindberg 1992; Stoddard 1994).

One classification scheme in particular has helped organize thinking about nitrogen retention by classifying forest systems based on stages of nitrogen saturation (i.e., the extent to which the system is saturated with nitrogen; the more saturated a system, the more likely to leach nitrogen) (Stoddard 1994). The classifications range from Stage 0, where forest nitrogen transformations are dominated by plant and microbial assimilation (uptake) with little or no NO_3 export from the watershed during the growing season, to Stage 3, where nitrogen deposition is well in excess of assimilation and has reduced plant and microbial assimilation capacities resulting in greater export of atmospheric nitrogen as well as losses from mineralization of soil organic nitrogen. Study sites in the southern portions of the Chesapeake Bay watershed generally fall into the low nitrate export classification (Stages 0-1), while the northern portions have generally high to medium export classifications (Stages 1-2).

Total Loadings Estimates Using A Mass Balance Approach. The role of atmospheric transport as an important path for nitrogen deposition to estuarine areas was first publicized in 1988 (Fisher et al. 1988). Based on a mass balance analysis using 1984 hydrology data, the authors estimated that one-third of the nitrogen entering the Chesapeake Bay is deposited from the atmosphere. Several subsequent efforts (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988) to quantify atmospheric nitrogen loadings to Chesapeake Bay produced "best-estimate loadings" ranging from 25 percent (Fisher and Oppenheimer 1991) to about 33 percent (Hinga et al. 1991) of the total nitrogen loads to the Chesapeake. (A discussion of the uncertainties in a mass balance approach from one of these studies is presented in the sidebar on the next page.)

The approach taken in these mass balance studies can be divided into two components: (1) estimating wet and dry deposition; and (2) estimating nitrogen retention. A central difficulty in mass balance studies is the use of average land use values of nitrogen retention. Nitrogen retention assumptions used in three of the Chesapeake Bay studies are presented in Table IV-8.

TABLE IV-8
Nitrogen Retention Assumptions Used in Chesapeake Bay Loading Studies
(as a percentage of nitrogen loading)

Land Use	Tyler 1988	Fisher and Oppenheimer 1991[a]	Hinga et al. 1991[a]
Forest	95.2-100.0	80.0 (51.0-100.0)	80.0 (25.0-95.0)
Pasture	93.7-99.96	70.0 (51.0-90.0)	80.0 (25.0-95.0)
Cropland	76.0-99.97	70.0	60.0 (45.0-75.0)
Residential	62.0-95.3	35.0 (0.0-70.0)	25.0 (10.0-50.0)

[a] Numbers in parentheses indicate range tested.

Assembling an adequate understanding of long-term behavior when the processes involved are fundamentally episodic is another major challenge of contemporary models. Some studies indicate that the majority of the atmospheric wet deposition occurs during a few episodes (Dana and Slinn 1988; Fowler and Cape 1984), such that the wet-deposited nitrogen (as well as previously dry-deposited nitrogen) is deposited directly to, or flows quickly into, the surface waters without intermediate reduction in concentration. Despite these difficulties, mass balance studies provide a good first-order estimate of nitrogen loading to Chesapeake Bay.

Experimental manipulation at the watershed scale is being conducted at a few U.S. locations (Kahl et al. 1993; Norton et al. 1994). Work from these sites is providing information on the cycling of nitrogen in forested catchments and is fully supportive of the conclusion that atmospheric deposition contributes to nitrogen loading of coastal waters through the export of atmospherically derived nitrogen. Results of these long-term experiments are just beginning to be published. An example is the Bear Brook watershed in Maine. Divided into treatment and control catchments, the treated catchment received increased nitrogen loading in the form of labeled ammonia. The treated watershed response was an immediate increase in stream nitrate export (Norton et al. 1994; Uddameri et al. 1995).

Inherent Uncertainties in Mass Balance Approach

"It would not be difficult to make the [mass balance] calculations appear more elegant by subdividing the watersheds into more land use types, using a detailed data base of land uses, assembling more detailed lists of point source and agricultural inputs, and using some technique for contouring deposition over the watershed. None of these approaches are likely to make better calculations. More precise and reliable estimates of the magnitudes of inputs of atmospherically-deposited nitrogen to coastal waters will require significant advances in the understanding of many processes responsible for the behavior of nitrogen in terrestrial ecosystems and in rivers and streams."

Source: Hinga et al. 1991.

Total Loadings Estimates Using the Chesapeake Watershed and Estuary Models. The Chesapeake Watershed Model (discussed in more detail below) is one approach to disaggregating the separate components of terrestrial and river nitrogen dynamics in the basin, along with the temporal effects of high loading during rainfall events. The estimate of atmospheric nitrogen loads for both direct and indirect deposition is 27 percent of the annual nitrogen load delivered to the Chesapeake Bay. This estimate was developed using the 1992 Watershed Model (Linker et al. 1993) and is consistent with the range reported by Chesapeake Bay mass balance studies (i.e., 25 to 33 percent). Further refinements are being made to the Watershed Model and an update of the estimate of atmospheric deposition is expected by September 1997.

To estimate wet deposition, the Chesapeake Bay Program combined output from a regression model developed from NADP weekly and daily precipitation chemistry measurements with data from the NOAA rainfall network. This approach yields daily estimates of rainfall to 74 sub-basins of the Chesapeake Bay watershed. Dry deposition was assumed to be equal to wet deposition for over-land areas and 44 percent of wet deposition for over-water areas. Indirect atmospheric loadings from the over-land portion of the watershed were estimated using the Chesapeake Bay Program Watershed Model.

The estimate of a 27 percent contribution of atmospheric deposition to total nitrogen loadings to the Chesapeake Bay falls within the range reported for other major eastern and Gulf coast estuaries, which are discussed in Section IV.D and summarized in Table IV-11 in that section.

MODELING THE CHESAPEAKE BAY'S WATERSHED AND ESTUARY

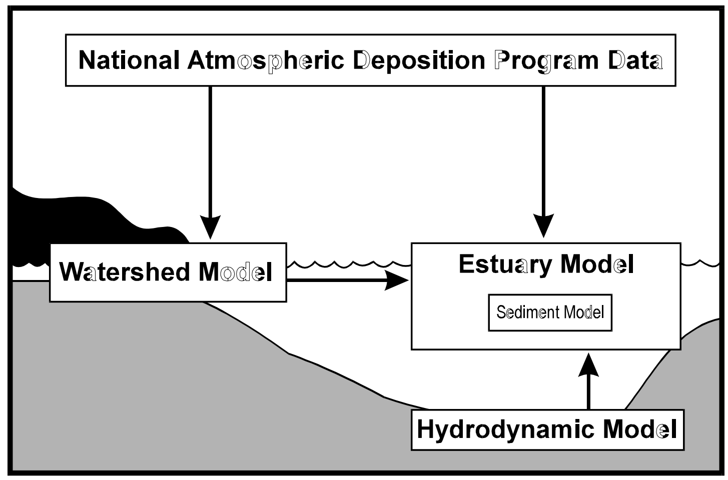
Water quality models of the Chesapeake Bay's watershed and estuary have been in use since the mid-1980s (CBP 1987; Donigian et al. 1991; Hartigan 1983; Thomann et al. 1994). The 1987 Bay Agreement's 40 percent nutrient reduction goal was based, in large part, on findings from these models.

The first model of the Bay watershed was completed in 1982 and provided a basin-by-basin assessment of the relative importance of point source and nonpoint source controls of nutrients (NVPDC 1983). Subsequent refinements of the Watershed Model established the importance of animal waste management in the watershed (Donigian et al. 1991), the delivery to the Bay of atmospheric deposition loads from the watershed (Donigian et al. 1994), and the development of tributary allocation loads of nitrogen and phosphorus to achieve the 40 percent nutrient reduction goal (Thomann et al. 1994).

In a parallel effort, the first model of the Chesapeake estuary was completed in 1987 to evaluate the impact of nutrient reduction scenarios on the Bay's dissolved oxygen concentrations (CBP 1987). Results from the steady-state Estuary Model indicated that a 40 percent reduction in nutrient loads would significantly reduce anoxia (dissolved oxygen concentrations less than 1 mg/L) in the Bay mainstem during average summer (June-September) conditions (CBP 1988). The 40 percent controllable nutrient reduction goal, under the 1987 Bay Agreement, was based in large part on these findings.

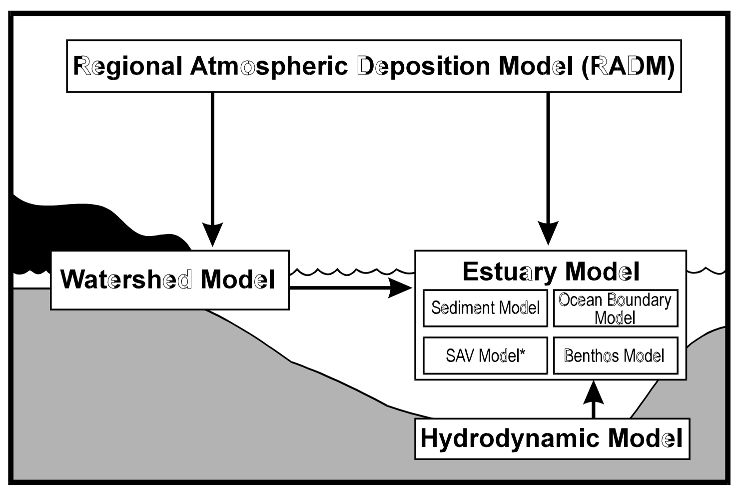
A reevaluation of the Bay's nutrient reduction goal and a review of the progress made in reducing nutrients was scheduled for 1992. In advance of this reevaluation, researchers began refining and integrating the Watershed and Estuary Models (Figure IV-14). For example, the Watershed Model was updated with greater detail of agricultural and atmospheric sources and was linked to the Estuary Model (Donigian et al. 1994). Providing a predictive framework for determining nutrient loads delivered to the tidal Bay under different source reduction scenarios, the Watershed Model simulated delivered nutrient loads with changes in land use practices and levels of wastewater treatment (Thomann et al. 1994). The Estuary Model was upgraded to add a sediment processes model and a hydrodynamic model, and was linked with the Watershed Model to accept Watershed Model nutrient loads as data input (Cerco and Cole 1994; DiToro and Fitzpatrick 1993; Johnson et al. 1991).

FIGURE IV-14
Watershed and Estuary Model Integration



The integrated Watershed and Estuary Model of the Chesapeake Bay was used to estimate water quality improvements that would be realized upon reaching the Bay Agreement goal of reducing controllable nutrients by 40 percent. Through the application of these models, the Bay Program partners established the Bay Agreement tributary nutrient allocations of nitrogen and phosphorus to be achieved by the year 2000 and maintained thereafter.

FIGURE IV-15
Integrated Model Improvements



* SAV = submerged aquatic vegetation

While this initial integrated model (Figure IV-14) could simulate the effects of atmospheric nitrogen deposition on water quality, it could not project the ultimate influence of changes in total nitrogen loadings (i.e., including loadings other than from the air) to the Bay. To provide for this predictive capacity, the Chesapeake Bay Program recently configured the Bay Watershed Model to accept daily atmospheric loadings by land use category (i.e., forest, pasture, cropland, and urban) (Linker and Thomann 1996). The Bay Watershed Model can now simulate the transport of increased or decreased atmospheric loadings to the Bay tidal waters along with nutrients from other land-based point and nonpoint sources. The Estuary Model is being upgraded to simulate basic ecosystem processes of submerged aquatic vegetation (SAV), benthic microorganisms, and major zooplankton groups. In addition, EPA's RADM is being directly linked to the Watershed and Estuary models. This new integrated model, functionally linking the airshed, watershed, estuary, and ecosystem, is expected to be completed in mid-1997. With these refinements, the integrated model (Figure IV-15) will simulate and evaluate the overall loads of controllable and uncontrollable nitrogen from the surrounding airshed and watershed, and the impact of these loads on the ecosystem. This model will be one of the tools used by Chesapeake Bay Program state and federal managers to formulate additional nitrogen reduction steps needed to achieve the 40 percent reduction goal and maintain the cap on nutrient loadings after the year 2000.

WATER QUALITY BENEFITS FROM REDUCING NITROGEN EMISSIONS

Using the Watershed Model, several scenarios were developed to examine the effectiveness of air emission controls compared to traditional point source and nonpoint source controls on the delivery of nutrient loads to the Chesapeake Bay. The Watershed Model scenarios were:

- *Base Case Scenario*: This scenario represents the base year 1985 loads to the Chesapeake Bay.

- *Bay Agreement Scenario*: This scenario represents the 40 percent controllable nutrient load reduction to be achieved by the year 2000 (as discussed under Nutrient Reduction Strategy in this section).

- *Bay Agreement plus CAA Scenario*: The scenario represents the controls on point and nonpoint source loads through the Bay Agreement, plus the atmospheric load reductions expected under full implementation of the CAA titles I (reductions in ground level ozone), II (mobile sources), and IV (utility sources).

- *Bay Agreement plus Ozone Transport Commission (OTC) Scenario*: The scenario evaluates reductions from the controls on point and nonpoint source loads through the Bay Agreement, plus the effects of implementation of CAA titles I, II and IV, as well as additional nitrogen reductions to reduce ground level ozone in the mid-Atlantic and New England metropolitan regions as called for by the Ozone Transport Commission.

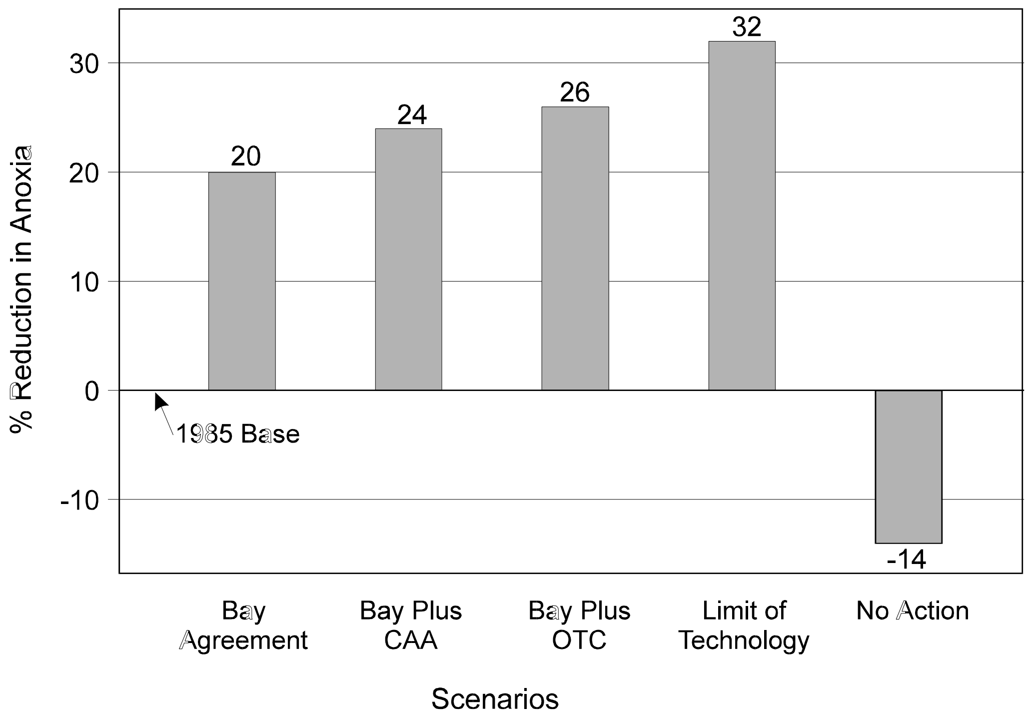
- *Limit of Technology Scenario*: This scenario estimates the nutrient reductions that may be realized using the current practical limit of point and nonpoint source control technologies, including conservation tillage for all cropland implemented; the Conservation Reserve program fully implemented; nutrient management, animal waste controls, and pasture stabilization systems implemented where needed; a 20 percent reduction in urban loads; and point source effluent controlled to a level of 0.075 mg/L total phosphorus and 3.0 mg/L total nitrogen. This scenario is significant because it determines the limit of currently feasible control measures.

- *No Action Scenario*: This scenario represents loads that would occur in the year 2000 given population growth and projected changes in land use. The controls in place in 1985 were applied to the year 2000 point source flows and land use, representing the loading conditions without the nutrient reductions stipulated in the Bay Agreement.

These reduction scenarios are part of an effort to evaluate options for achieving the 40 percent nutrient reduction goal. Land-based nonpoint source and wastewater treatment facility-based point source reduction actions, planned for implementation in many Chesapeake tributary watersheds, are approaching the limits of technology. Options for reductions in air emissions are being explored for maintaining the targeted 60 percent nutrient loadings cap beyond the year 2000 in the face of a growing population and resultant development in the watershed. Different options will have different costs and effectiveness with regard to water quality improvements, and a range of options should be evaluated to find the best approach.

The water quality improvement from the expected reduction in nitrogen emissions under each scenario are shown in Figure IV-16. The improvement in water quality reflects the estimated reductions in Bay bottom waters having no dissolved oxygen (i.e., reduction in Chesapeake anoxia or "dead waters"). Decreased nitrogen loadings will result in decreased water column nitrogen which will, in turn, decrease the growth of algae and improve the level of light penetration necessary to support the critically important SAV (Batiuk et al. 1992; Dennison et al. 1993; Thomann et al. 1994).

FIGURE IV-16
Reductions in Anoxia Under Nutrient Reduction Scenarios



The CAA and OTC scenarios indicate that these controls provide for nitrogen loading reductions and water quality improvements above and beyond those provided by implementation of the Chesapeake Bay Agreement commitment of 40 percent reduction in controllable nitrogen. Relative reductions from controls on sources of atmospheric deposition vary by tributary basin, with the more sensitive tributaries being the Susquehanna and the Potomac. These basins receive the highest deposition loads in the Chesapeake watershed and are among the most responsive to reductions in atmospheric deposition.

Though the differences between scenarios in percent reductions in anoxia might seem small, air emission controls could account for a fifth and a third of the baywide nitrogen load reduction goal through CAA implementation and OTC reductions, respectively. Such reductions could make achieving the 40 percent reduction target more feasible, and make maintaining a cap on any further increases in nutrient loadings beyond 2000 possible. These additional reductions are especially important in the face of increasing population and watershed development that studies predict will increase the significance of atmospheric deposition as a source of nitrogen loadings to the Chesapeake Bay in the coming decades (Fisher et al. 1988; Pechan 1991).

AREAS OF UNCERTAINTY AND WORK UNDERWAY

Nitrogen retention, the relative loadings of ammonia and organic nitrogen, and dry deposition to water surfaces are a few of the remaining areas of significant uncertainty in estimating atmospheric nitrogen loads. Several specific examples of areas of uncertainty are discussed below.

- ***Nitrogen retention*** within watersheds makes a big difference in the proportion of the atmospheric contribution to nitrogen loading to the Bay. Different retention assumptions in mass balance analyses lead to an uncertainty in the estimate of this contribution by more than a factor of two.

- ***Ammonia and organic nitrogen*** contribute a large portion of nitrogen deposition, perhaps more than 25 percent of the total atmospheric nitrogen load. However, it is unknown to what degree their sources are controllable, and they may be changing with time. For example, airborne ammonia emissions from agricultural animal operations could increase.

- Estimates of the relative contribution of ***dry deposition*** to the total atmospheric deposition loadings range from 25 to 63 percent (Duce et al. 1991; Levy and Moxim 1987; Logan 1983; Lovett and Lindberg 1986; Sirois and Barrie 1988; Walchek and Chang 1987). Faced with this wide range of estimates, many investigators choose to set the dry deposition loading equal to the measured wet deposition loading. This assumption is known to be questionable. While site-specific data to refine the estimate are lacking, recent evaluations indicate that dry deposition to tidal water surfaces is about 25 percent of wet deposition to tidal water surfaces (Luke and Valigura 1997).

- ***1990 baseline emission estimates*** continue to be refined. Estimates of emissions from off-road vehicles have been significantly improved. Ship emissions in harbors are suspected to be significantly underestimated (Booz-Allen & Hamilton 1991). While emissions from these sources are not large in the aggregate, they occur close to the Bay tidal surface waters, and thus have an influence greater than their national fractions would imply. Nitrogen emissions from on-road vehicles continue to be a source of uncertainty.

- ***Particulate nitrate*** (which has a low deposition velocity) and ***nitric acid*** (which has a high deposition velocity) are currently indistinguishable by RADM, leading to modeling uncertainty.

- The contribution of atmospheric nitrogen deposition to ***offshore ocean waters*** has not yet been characterized. The ocean waters exchange with waters of the Chesapeake Bay and thus may be a source, or a sink, of nitrogen loads to the Bay.

To reduce existing uncertainties in atmospheric loadings estimates, the Chesapeake Bay Program convened a workshop in June 1994, inviting scientists and managers with expertise and experience in understanding or managing atmospheric deposition. The challenge given to attendees was to construct a list of practical studies that would make the greatest impact on reducing the current uncertainty in estimates of the contribution of atmospheric deposition to declining aquatic ecosystem health. The resulting list (CBP 1995a) is summarized below:

♦ *Priority 1*: Conduct intensive, coordinated, and integrated monitoring studies at special locations within the watershed that characterize wet deposition, dry deposition, and local catchment area.

♦ *Priority 2*: Improve existing atmospheric models (e.g., reduce grid size, account for the effect of mountains).

♦ *Priority 3*: Improve models of chemical retention in watersheds.

♦ *Priority 4*: Improve emissions inventories and projections.

♦ *Priority 5*: Conduct measurements to extend vertical and spatial meteorological and chemical concentration coverage in models.

♦ *Priority 6*: Establish an extensive array of less intensive measurement stations to improve spatial resolution for selected variables.

To improve the cross-media modeling capabilities and to reduce existing sources of uncertainty in atmospheric deposition loadings estimates, the following work is underway through cooperation between EPA, state and federal agencies, and universities:

- Measuring the concentration of atmospheric organic nitrogen within the Chesapeake Bay watershed;

- Measuring dry deposition of nitrate to tidal surface waters of the Bay;

- Investigating the atmospheric deposition of dissolved organic nitrogen and its isotopic composition (delta (δ) ^{15}N);

- Linking daily atmospheric deposition and resultant nitrogen runoff from pasture, forested, and urban lands within the Chesapeake Bay Watershed Model;

- Decreasing the grid size of RADM across the Bay watershed to increase the spatial resolution and improve the resultant model scenario output; and

- Linking RADM with the Watershed, Estuary, and Water Quality models, including simulation of atmospheric deposition to offshore ocean waters and exchange of the ocean waters with Chesapeake Bay waters.

The result of this and other work will become part of the integrated model of the Bay's airshed, watershed, estuary, and ecosystem (discussed above), which is expected to be completed in early

1997. A series of management scenarios, similar to the modeling scenarios discussed above, are also expected to be completed in 1997 to examine the most feasible and cost-effective combination of point source, nonpoint source, and air deposition reductions to meet the Chesapeake Bay Agreement commitment to cap nutrient loadings in 2000 at 60 percent of 1985 controllable base loadings and to ultimately restore the water quality conditions necessary to fully support the Bay's invaluable living resources.

Toxic Contaminant Deposition to the Chesapeake Bay

CHESAPEAKE BAY BASINWIDE TOXICS STRATEGY

The 1987 Chesapeake Bay Agreement committed the signatories to "develop, adopt, and begin implementation of a basinwide strategy to achieve a reduction of toxics consistent with the Clean Water Act of 1987, which will ensure protection of human health and living resources" (Chesapeake Executive Council 1987). The resultant strategy, adopted in 1989, initiated a multi-jurisdictional effort to more accurately define the nature, extent, and magnitude of Chesapeake Bay chemical contaminant problems and to initiate specific chemical contaminant reduction and prevention actions (Chesapeake Executive Council 1989). Building on a two-year reevaluation of the strategy and increased understanding of the nature of the Bay's toxics problems, a revised, farther-reaching strategy was adopted in 1994. The 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy* recognized the contribution of atmospheric deposition as a significant source of chemical contaminant loadings to the Bay. ***Within the basinwide strategy, the signatories committed to establishing a more comprehensive loadings baseline and setting an atmospheric deposition loading reduction target to be achieved over the next decade*** (Chesapeake Executive Council 1994).

In 1991, the Chesapeake Bay Program adopted its first Chesapeake Bay Toxics of Concern list, principally to identify and provide concise documentation on chemical contaminants that adversely affect the Bay or have a reasonable potential to do so. This list provided Chesapeake Bay region resource managers and regulators with a baywide consensus of priority chemicals and the information necessary to target these chemical contaminants for strengthened regulatory and prevention actions or additional research, monitoring, and assessment. Based on ambient concentrations of chemical contaminants and aquatic toxicity data, the toxic pollutants that represented immediate or potential threats to the Chesapeake Bay system were identified. The Toxics of Concern list (see sidebar) includes several pollutants of concern for atmospheric deposition to the Great Waters (cadmium, chlordane, lead, mercury, PAHs, and PCBs). In addition, a Chemicals of Potential Concern list was identified for the Chesapeake Bay (see sidebar) and includes two pollutants of concern for the Great Waters (dieldrin and toxaphene). Clear evidence is lacking that the contaminants on the Chesapeake Bay list of Chemicals of Potential Concern actually cause or have reasonable potential to cause adverse effects in the environment, but the Chesapeake Bay Program believes these

Chesapeake Bay Toxics of Concern

Toxics of Concern: atrazine, benz(a)anthracene,[a] benzo(a)pyrene,[a] cadmium, chlordane, chromium, chrysene,[a] copper, fluoranthene,[a] lead, mercury, napthalene,[a] polychlorinated biphenyls (PCBs), and tributyltin.

Chemicals of Potential Concern: alachlor, aldrin, arsenic, dieldrin, fenvalerate, metolachlor, permethrin, toxaphene, and zinc.

[a] A polycyclic aromatic hydrocarbon (PAH).

chemicals warrant enough concern to be carefully monitored and tracked. For example, a number of the chemicals listed as being a potential concern are either banned or restricted pesticides that have residues still remaining in the ecosystem at elevated levels but below thresholds of concern; others are chemicals of increasing concern due to use patterns or potential for toxicity to Bay resources.

In response to a commitment within the 1994 Basinwide Toxics Strategy, the Toxics of Concern list is currently being evaluated and revised using a risk-based chemical ranking system incorporating source, fate, and exposure/effects ranking factors. In-depth analyses of the top-ranked chemicals will lead to the selection of a revised Toxics of Concern list in 1997.

CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC DEPOSITION STUDIES

Studies conducted in the southern Chesapeake Bay in the early 1980s suggest that the atmosphere is a significant source of organic contaminants to the Bay, such as organic carbon (Velinsky et al. 1986) and anthropogenic hydrocarbons, including PAHs (Webber 1993). While these earlier studies demonstrated the potential importance of the atmosphere in supplying contaminants to the Chesapeake Bay, they were limited by their methodologies (i.e., bulk deposition sampling, which is imprecise) and their relatively limited temporal and spatial scope. An assessment of the extent of toxic contamination in the Bay is presented in the sidebar.

To further explore the issue of atmospheric loadings of toxic contaminants to the Bay, the Chesapeake Bay Atmospheric Deposition Study (CBADS) network was established by a team of scientists from the University of Maryland, Virginia Institute of Marine Sciences, University of Delaware, and Old Dominion University. The primary objective of the CBADS network was to provide the best estimate of total annual atmospheric loadings of a variety of trace elements and organic contaminants directly to the surface waters of the Chesapeake Bay. Because accurate estimates of baywide annual loadings require characterizing the spatial and temporal variability in contaminant loads in the atmosphere and in depositional fluxes to the Bay, CBADS collected data to help characterize this variability.

Extent of Toxic Contamination in Chesapeake Bay

Prior to adoption of the 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy*, the Chesapeake Bay Program conducted a two-year, in-depth evaluation of the nature, extent, and magnitude of toxic contaminant-related problems within the Chesapeake Bay. Through the evaluation, no evidence was found of severe baywide impacts from chemical contamination, unlike other problems facing the Chesapeake Bay, such as the impacts from excess nutrients. The Program did document severe, *localized* toxicity problems, adverse effects from chemical contamination on aquatic organisms in areas previously considered unaffected, and widespread low levels of chemical contamination in all Bay habitats sampled.

Existing state and federal regulatory and management programs continue to reduce the input of potentially toxic chemicals to the Chesapeake Bay. Measured concentrations of many of these chemical contaminants in the Bay's bottom sediments, shellfish, fish, and wildlife have also generally declined, although elevated levels occur in several industrialized areas and some increasing trends have been observed. Progress in reducing the point sources of these chemical contaminants is offset by significant nonpoint source inputs of chemical contaminants (e.g., urban stormwater runoff, atmospheric deposition) from increasing development and urbanization of the Bay watershed.

Based on previous studies of wet deposition in the region (e.g., Tyler 1988) and given the resources available for the network, three non-urban shoreline locations -- Wye Institute and Elms Institute, Maryland, and Haven Beach, Virginia -- were selected and sampled from 1990 to 1993. These three sampling site locations, as well as other monitoring sites around Chesapeake Bay, are shown in Figure IV-17. In establishing this initial network, the influence of urban areas was purposely avoided by locating the sites more than 50 kilometers from metropolitan areas (similar to the initial non-urban stations for the Great Lakes deposition network, the Integrated Atmospheric Deposition Network (IADN)). By minimizing possible urban influences, the resulting CBADS loading estimates are considered to be minimum values.

CBADS evaluated atmospheric loadings directly to the Bay only. Although it is most likely that some fraction of the toxic contaminants deposited from the atmosphere to the watershed of the Chesapeake Bay are ultimately transported to the surface waters, this study did not attempt to characterize indirect loadings for two main reasons:

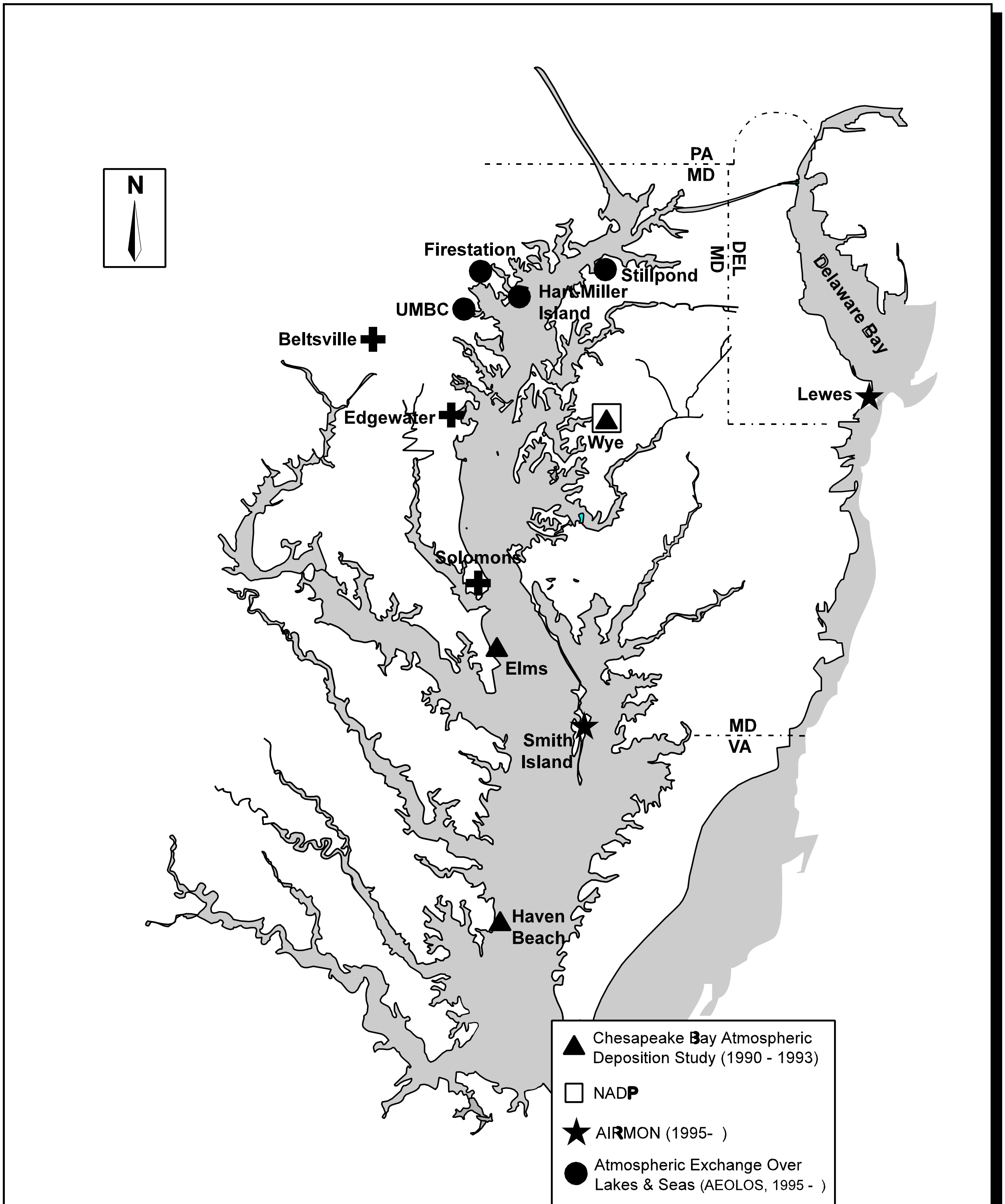
- Because deposition to the various land surfaces is likely much different than that to the water surface, fluxes measured at the shore-based stations cannot be extrapolated with confidence to the watershed; and

- The large uncertainty in the understanding of the fate of materials deposited to the land surface (i.e., the fraction transmitted to the receiving water) precludes the simple estimation of the indirect atmospheric loading of contaminants to the Chesapeake Bay in more than a rough estimate.

The CBADS data on concentrations in air, concentrations in precipitation, and wet and dry aerosol depositional fluxes are presented in Baker et al. (1997) and are summarized below. These data were collected for two groups of contaminants: trace metals (aluminum, arsenic, cadmium, chromium, copper, iron, manganese, nickel, lead, selenium, and zinc); and organic contaminants (14 different PAHs and total PCBs). Cadmium, lead, PAHs, and PCBs are pollutants of concern for atmospheric deposition to the Great Waters. Mercury data are being collected but have not yet been compiled, and data on other Great Waters pollutants of concern were not collected. For detailed results, a discussion of sampling methods, or a description of the limitations of the study, refer to Baker et al. (1997).

Concentrations in Air. Air concentrations for trace metals were determined by measuring the elemental composition of aerosol particles less than 10 μm in diameter. The elemental composition was dominated by the crustal elements aluminum and iron, as well as sulfur (in the form of sulfate). During 1991 and 1992, concentrations averaged over the three sampling sites were 116, 111, and 2,123 ng/m^3 for aluminum, iron, and sulfur, respectively. Trace element concentrations averaged over the same period ranged from 0.16 ng/m^3 for cadmium to 12.6 ng/m^3 for zinc, with lead averaging 3.88 ng/m^3. The fraction of each element derived from non-crustal (e.g., combustion) sources was estimated based on the average concentration of elements in the Earth's crust (Turekian and Wedepohl 1961), and assuming all of the measured aluminum associated with aerosol particles is derived from erosion of soils. In the Chesapeake Bay region, non-crustal sources supply greater than 95 percent of most of the elements measured on aerosol particles (Wu et al. 1994). Arsenic, cadmium, lead, sulfur, and selenium are almost exclusively non-crustal, and are likely introduced into the atmosphere as a result of combustion of fossil fuels and incineration of municipal wastes.

FIGURE IV-17
Sampling Locations for
Chesapeake Bay Toxic Contaminant Atmospheric Deposition Studies



Source: Baker et al. 1996

The measured concentrations of trace elements were generally within a factor of two among the three sampling sites. On an average annual basis, concentrations slightly decrease from north (Wye) to south (Haven Beach), except sulfur, which is 15 percent higher at Elms (1991 and 1992) and Haven Beach (1992) than at the Wye site. The general decreasing trends observed from north to south, along with increasing sulfate, may indicate higher levels of low sulfur combustion sources (e.g., incinerators, vehicles) in the northern reaches of the Chesapeake Bay. In general, the spatial variability in the atmospheric concentrations of trace elements between sites (from north to south) is substantially lower than corresponding temporal trends.

Semivolatile organic chemicals exist in the atmosphere as gases and also are associated with aerosol particles (Pankow 1987). In this study, baywide annual average concentrations of PAHs in air ranged from 16 ng/m^3 for dibenz[*a,h*]anthracene to 2,590 ng/m^3 for phenanthrene. Atmospheric concentrations were quite variable with individual measurements ranging from one-tenth to ten times the annual average concentrations. These variations likely result from sampling air masses coming from differing directions, from changes in local and regional emissions, and from differences in atmospheric degradation and deposition rates. These variations show a seasonal pattern. For example, increased concentrations of gas-phase PAHs, such as pyrene, during the summer months may reflect both higher temperatures (i.e., enhanced volatilization) and increased coal and oil combustion to meet the electricity demand of air conditioning. Increases in the atmosphere of particulate PAHs, such as benzo[*a*]pyrene, may result from local burning of yard wastes and of wood for home heating. Some variation in atmospheric levels of organic chemicals may result from the efficient removal of particulate PAHs by precipitation (Poster and Baker 1996a, 1996b). In general, the magnitude of fluctuations in atmospheric levels of organic chemicals is larger than the corresponding variations in trace elements and sulfur discussed above, suggesting the importance of local combustion sources. Air concentration data were not available for PCBs.

Concentrations in Precipitation. The overall volume-weighted mean concentrations of trace elements in precipitation collected at the three sites range from 0.03 μg/L for cadmium at Elms to 14.6 μg/L for iron at Haven Beach. For lead, the range of overall volume-weighted mean concentrations was between 0.42 and 0.52 μg/L at the three sites. The relative proportion of trace elements in precipitation is nearly identical to that in the Chesapeake Bay aerosol particles, confirming that aerosol scavenging is the source of trace metals to wet deposition. Trace metal wet depositional fluxes are highly variable, changing more than ten-fold between consecutive months with little apparent seasonal dependence. This variability, which was similar at each of the three sites, results not only from fluctuations in the atmospheric inventories of trace metals, but also from changes in the amount of precipitation. On an annual basis, the volume-weighted mean concentrations of most trace metals did not systematically vary between the summer of 1990 and fall of 1993, again suggesting that these rural sites were most strongly influenced by the same regional background signal throughout the study.

While individual precipitation events result in spikes in trace metal deposition at one site that are not observed at the other two locations, these isolated events average out over the year. Annual volume-weighted mean concentrations of trace metals in precipitation are generally within a factor of two among the three sites, with no clear systematic spatial trend observed for all metals.

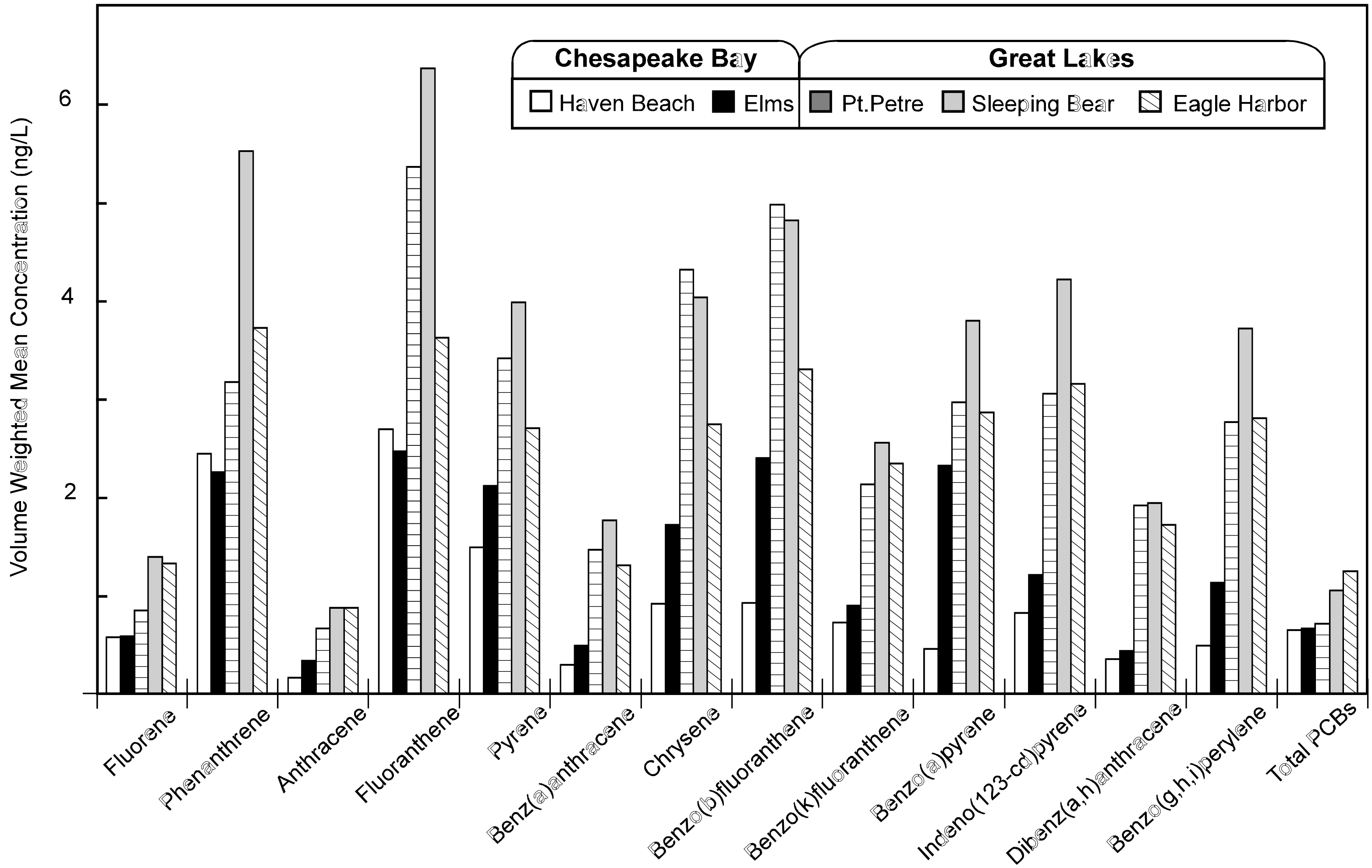
Overall volume-weighted mean concentrations of PAHs in precipitation ranged from 0.3 ng/L for anthracene and dibenz[*a,h*]anthracene at the Havens Beach site to 9 ng/L for pyrene at the Elms site. Volume-weighted mean concentrations for total PCBs were 1.1 and 0.9 ng/L at the Elms and Haven Beach sites (collected from June 1990 through September 1993), and 0.35 ng/L at the Wye site (collected from January to September 1993). As was observed for trace metals, wet depositional fluxes of organic contaminants varied considerably with time, and were dominated by episodic spikes at each location. Extremely high concentrations of some analytes, including pyrene, that were measured in both air and precipitation at the Elms site in the summer of 1990, may have resulted from local vegetation burning. Unlike PAH levels in the atmosphere, concentrations in precipitation did not systematically vary with season. The enrichment at the Elms site is especially pronounced for higher molecular weight PAHs, suggesting a local combustion source (e.g., wood burning for residential heating).

Concentrations of organic contaminants in precipitation measured in this study are consistently lower than those observed in the Great Lakes region (see Figure IV-18). For example, PAH concentrations in precipitation at the Chesapeake Bay sites are one-third to one-half as high as at the three IADN sites located at rural, shoreline sites on Lakes Ontario, Michigan, and Superior (Gatz et al. 1994). In contrast, levels of the same PAHs in the air over Chesapeake Bay are equal to or perhaps higher than those measured over the Great Lakes. Whether the apparent enrichment in PAHs in Great Lakes precipitation relative to that in the Chesapeake Bay region, as shown in Figure IV-18, is due to more efficient scavenging by precipitation in the colder, relatively drier Great Lakes region, or simply reflects methodological differences between the two networks, is unclear.

Wet and Dry Aerosol Depositional Fluxes. Using CBADS data, researchers calculated depositional fluxes (see Baker et al. (1997) for methodologies). Because these "depositional fluxes" are actually gross fluxes directly to the water surface and do not account for net gas exchange across the air-water interface, the term "deposition rates" is used in the remainder of this discussion in place of "depositional fluxes" to be consistent with the rest of the report in the use of the term "flux."

Total annual deposition rates in 1992 ranged from 0.07 mg/m^2 for cadmium at the Wye site to 121 mg/m^2 for aluminum at the Elms site; the highest annual deposition rate for lead was 1.34 mg/m^2 at the Wye site. Not surprisingly, dry aerosol deposition comprises the majority of the total deposition rate for the soil elements aluminum and iron, which occur on coarse particles. Wet deposition contributes between one-third and one-half of the total depositional rate of the remaining trace elements. Naturally, spatial trends in total deposition result from variation in precipitation chemistry and amount, and the trace element inventories associated with aerosol particles (given the considerable uncertainty in the dry aerosol deposition calculation, the same deposition velocity was used at each site). Although a distinct north to south trend in precipitation amount occurred in 1992 (100, 107, and 122 cm, respectively), total annual deposition rates were remarkably similar among the three stations. Total deposition rates were also very similar between years, again indicating that the relatively rare spikes in concentration are dampened against the chronic regional background signal.

FIGURE IV-18
Comparison of 13 PAHs and Total PCBs in Precipitation (1992) from Chesapeake Bay and Great Lakes Sampling Sites



Source: Baker et al. 1996 (Chesapeake Bay) and Gatz et al. 1994 (Great Lakes).

For PAHs, total annual deposition rates for 1992 range from 0.2 μg/m^2 for anthracene at the Wye site to 10.8 μg/m^2 for benzo[*b*]fluorathene at the Elms site.[12] Both wet deposition and dry aerosol deposition contribute to total PAH deposition, with dry aerosol deposition becoming relatively more important for the higher molecular weight, less volatile compounds. Total deposition rates for PAHs decrease with time during this study, with the lowest rates occurring during the first nine months of 1993. While some of this decrease is attributed to beginning with anomalously high measurement in the summer of 1990, decreases in both wet and dry aerosol deposition rates continued between 1992 and 1993. The total annual deposition rate for total PCBs is about 3.5 μg/m^2, with approximately equal contribution from wet and dry aerosol deposition.[13]

Overall, total annual deposition rates for PAHs and PCBs are generally within 50 percent among the sites. Given the uncertainty in the dry aerosol deposition estimates, this percentage indicates little spatial variability when integrating over annual cycles. However, this study did not specifically address the possible influences of urban areas, such as the cities of Baltimore, Washington, and Norfolk, on atmospheric deposition, which may be important.

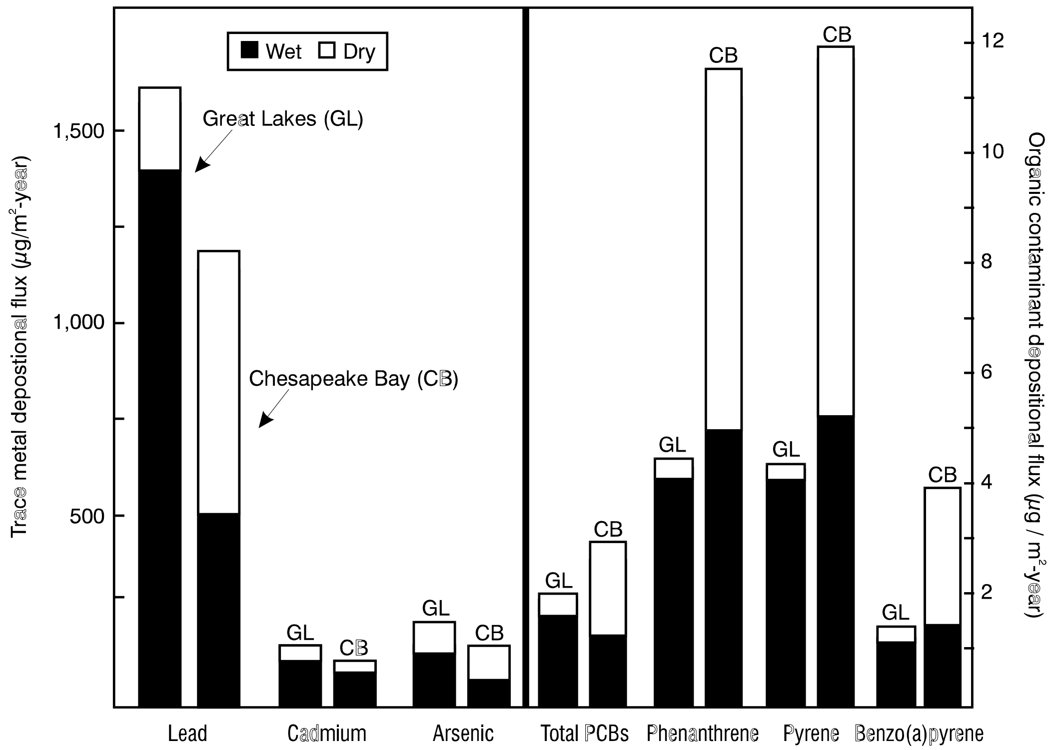
To place the atmospheric deposition rates calculated in this study in perspective, they are compared to similar estimates made for the Great Lakes region (Figure IV-19). Wet deposition rates for lead and arsenic are almost three times higher in the Great Lakes than in Chesapeake Bay, despite significantly less rainfall (80 versus 110 cm/year); wet deposition rates for cadmium are similar for both regions. Wet deposition rates for PAHs and total PCBs are fairly similar between the two regions, as higher concentrations in Great Lakes precipitation (see Figure IV-18) are offset by lower precipitation amounts. Dry aerosol deposition rate estimates are higher in the Chesapeake region, especially for organic contaminants.[14] In addition, measured aerosol-bound organic concentrations were generally higher than the values used in the Great Lakes dry aerosol deposition calculations (Eisenreich and Strachan 1992). Despite the differences, estimated atmospheric deposition rates are generally within a factor of two between the Chesapeake Bay and the Great Lakes regions, which, given the numerous opportunities for error in these measurements and calculations, is quite good agreement.

[12] Because the "fluxes" in this study are actually gross fluxes directly to the water surface, these data do not take into account exchange of gaseous organic contaminants across the air-water interface. Other recent studies have shown that this is the *dominant* atmospheric deposition process for semi-volatile organic contaminants, including PCBs (Baker and Eisenreich 1990; Achman et al. 1993) and low molecular weight PAHs (Nelson et al. 1995). In those studies, the net direction of exchange is often from the water to the air and diffusive gas exchange is large enough to offset wet deposition and dry aerosol deposition.

[13] Because aerosol particle-associated PCBs were present below analytical detection limits, estimates of PCB dry deposition were made using an aerosol PCB level calculated from the measured gaseous PCB concentration and the Junge-Pankow sorption model (Pankow 1987; Leister and Baker 1994).

[14] This is due, in part, to the choice of deposition velocities used for the two studies (0.2 cm/sec for all species in the Great Lakes, 0.26 cm/sec for trace elements and 0.49 cm/sec for organics in the Chesapeake Bay). All of these values are within the generally accepted range for dry aerosol deposition velocities.

FIGURE IV-19
Comparison of Chesapeake Bay and Great Lakes Atmospheric Depositional Fluxes



Source: Baker et al. 1996 (Chesapeake Bay) and Eisenreich and Strachan 1992 (Great Lakes).

CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC LOADINGS

To estimate the annual baywide loadings of trace elements and organic contaminants to the Bay, the annual site-specific wet and dry aerosol deposition rates were averaged and the two average rates were multiplied by the surface area of the Bay. Uncertainties in these loading estimates are likely, on the order of a factor of two, mainly due to the inability to estimate dry aerosol loadings on a finer temporal resolution.

Baywide atmospheric loadings of aluminum and iron are estimated at 1,340,000 and 799,000 kg/year, respectively (see Table IV-9). Loadings of trace elements range from 1,110 kg/year for cadmium to 49,400 kg/year for nickel; lead loadings are estimated at 12,500 kg/year. Loading estimates are generally similar for 1991 and 1992, except for nickel and zinc due to

TABLE IV-9
Annual Atmospheric Loadings of Trace Metals and Organic Contaminants to the Chesapeake Bay[a]

Pollutant	Wet Deposition (kg/year)	Dry Deposition (kg/year)	Total Deposition (kg/year)
Aluminum[b]	137,000	1,200,000	1,340,000
Arsenic	607	1,050	1,660
Cadmium	867	240	1,110
Chromium	1,026	2,030	3,060
Copper	5,575	3,620	9,200
Iron	132,800	666,000	799,000
Manganese	13,200	13,600	26,800
Nickel	7,185	6,160	13,300
Lead	5,440	7,080	12,500
Selenium	1,390	2,930	4,320
Zinc	26,000	22,800	49,400
Total PCBs	13	20	37
PAHs			
Anthracene	6	6	13
Benz(*a*)anthracene	9	34	44
Benzo[*a*]pyrene	17	36	53
Benzo[*b*]fluoranthene	36	98	134
Benzo[*e*]pyrene	21	67	88
Benzo[*ghi*]perylene	19	75	94
Benzo[*k*]fluoranthene	22	65	88
Chrysene	29	85	114
Dibenz[*ah*]anthracene	7	16	22
Fluoranthene	70	119	189
Fluorene	16	12	27
Indeno[123*cd*]pyrene	20	78	98
Phenanthrene	63	92	155
Pyrene	75	109	184

[a] To estimate annual baywide loadings of elements and organic contaminants to the entire Chesapeake Bay, the annual site-specific wet and dry aerosol fluxes were averaged and these two average fluxes were multiplied by the surface area of the Bay.
[b] Contribution of aluminum is considered to be entirely from natural sources (i.e., not emitted through human activities).

Source: Baker et al. 1997.

elevated wet deposition measured at the Haven Beach site in 1992. Loadings of PAHs range from 13 kg/year for anthracene to 189 kg/year for fluoranthene. Total PCB loadings are estimated to be 37 kg/year. Interestingly, for many of the pollutants in Table IV-10, wet deposition and dry aerosol depositional fluxes appear to decrease between 1991 and 1992. Whether this reflects a real inter-annual variation or simply results from aggregating measurements from different locations is unclear.

To place these loadings in perspective, they are compared in Table IV-10 to recent estimates of trace metal and organic contaminant loadings delivered to the Chesapeake Bay by the Susquehanna River (Conko 1995; Foster 1995; Godfrey et al. 1995). The Susquehanna River is the largest tributary of the Chesapeake Bay, supplying approximately 60 percent of the freshwater inflow to the estuary. Annual riverine loadings of dissolved and particulate trace metals and organic contaminants were determined by analyzing flow-weighted samples collected at Conowingo, Maryland, between February 1994 and January 1995 (Conko 1995; Foster 1995). Atmospheric deposition directly to the surface waters of the Chesapeake Bay supplies PAH loads that are comparable to or greater than the loads of dissolved PAHs delivered by the Susquehanna River (Table IV-10). Particulate-bound organic contaminants discharged from the river dominate the loading of PAHs, with a large contribution from the high sediment burden carried by the river during high flows. Dissolved total PCB loads from the river are approximately three times those from the atmosphere. Atmospheric depositional fluxes of several elements, including lead, cadmium, and chromium, are within a factor of two of the dissolved load from the Susquehanna River. Again, particulate metal loads from the river dominate over both dissolved riverine loads and atmospheric deposition.

While it is interesting to compare the relative importance of riverine and atmospheric sources of trace elements and organic contaminants to the Chesapeake Bay, the results should be carefully interpreted. While the Susquehanna River delivers large quantities of these substances to the Bay, much of this load is removed in the northern extreme of the Bay (Helz and Huggett 1987) and is delivered episodically during high river flows. Whether particulate-bound metals and organic contaminants are broken down in forms that can be taken up by aquatic organisms is quite unclear. In contrast, atmospheric deposition directly to the water's surface supplies these toxics directly to the water column, without any comparable zone of efficient removal. However, it has recently been suggested that combustion-derived PAHs associated with aerosol particles washed into the surface waters by precipitation also may not be broken down (McGroddy and Farrington 1995). Finally, the distinction between riverine and atmospheric loadings is not clear. Some fraction of the pollutant input from the tributaries results from deposition of atmospheric pollutants to the watershed, with subsequent transmission through the vegetation and soil layers into surface waters (i.e., indirect loading); however, this input cannot yet be quantified.

AREAS OF UNCERTAINTY AND WORK UNDERWAY

Building on the existing CAA requirements, the Chesapeake Bay Program's state and federal partners will focus their efforts on implementation of the Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy commitment to "establish a more complete baseline and source identification for atmospheric deposition...and set a reduction target from that baseline to be achieved over the next decade" (Chesapeake Executive Council 1994). However, there are several remaining areas of uncertainty to be addressed related to atmospheric deposition of toxic contaminants to Chesapeake Bay. Two significant ones are:

TABLE IV-10
Relative Importance of Sources of Trace Metals and Organic Contaminants to Chesapeake Bay

Pollutant	Susquehanna River Load (kg/year)[a]		Atmospheric Deposition Load (kg/year)[b]
	Dissolved	Particulate	
Aluminum	2,560,000	64,800,000	1,340,000
Arsenic	12,600	ND	1,660
Cadmium	2,130	26,700	1,110
Chromium	4,130	111,000	3,060
Copper	47,800	151,000	9,200
Iron	4,100,000	40,000,000	799,000
Manganese	3,290,000	1,530,000	26,800
Nickel	121,000	65,200	13,300
Lead	6,530	38,600	12,500
Zinc	77,900	360,000	49,400
Total PCBs	97	68	37
PAHs			
Benz[*a*]anthracene	12	364	44
Benzo[*a*]pyrene	5	436	53
Chrysene	15	316	114
Fluoranthene	108	1,020	189
Fluorene	37	85	27
Phenanthrene	63	388	155
Pyrene	104	925	184

[a] Annual loads entering the Chesapeake Bay via the Susquehanna River, measured at Conowingo, Maryland, between February 1994 and January 1995 by Foster (1995) for organic compounds and Conko (1995) for metals.
[b] Total atmospheric deposition loads directly to the surface of the Chesapeake Bay as measured by CBADS.

Source: Baker et al. 1997.

♦ *Dry Deposition*. Dry deposition is viewed as an important mechanism by which chemical contaminants are deposited onto the Bay's tidal surface waters and surrounding watershed. As is the case with nitrogen, there are no widely accepted techniques for direct measurement of dry deposition fluxes of metal or organic contaminants. Although no direct measurements of dry deposition directly to the Bay exist, depositional fluxes have been estimated based on a particle-size-dependent deposition velocity function applied to direct measurements of aerosol concentrations of metals and organic chemical contaminants. Given the absence of direct measures of dry deposition fluxes, there is much uncertainty associated with these loading estimates.

♦ *Transport through the Watershed*. Atmospheric deposition of a pollutant can be a direct input to the Bay surface waters or can be transported from the watershed by surface water and groundwater to the Bay. Transported loads are a component of the total fluvial (i.e., surface water) input from the watershed to the Bay. The degree of landscape retention for a given substance is related to the geomorphology, land use, basic hydrological characteristics unique for each watershed, and soil chemistry. Limited studies to date suggest that the degree of watershed throughput is relatively small (< 30 percent of the rate of rainfall volume). However, evidence to date suggests that watersheds serve as a "reservoir" for atmospherically deposited metals; organically bound metals are sequestered but may be episodically mobilized by acidic precipitation. Because of the relatively large watershed to open water surface area ratio typical of coastal plain estuaries such as Chesapeake Bay (15:1), recent estimates for nitrogen and trace elements suggest that the indirect atmospheric loading may be as significant as the direct input. Thus, while it has been possible to quantify direct atmospheric flux with a fair degree of confidence, one of the primary uncertainties associated with resolving the total atmospheric loading to Chesapeake Bay is in gauging the indirect loading as it relates to the watershed transmission/retention for the myriad of sub-basins (Valigura et al. 1995).

To further improve existing estimates of the relative atmospheric deposition contribution to total chemical contaminant loadings to Chesapeake Bay, the following work is underway. In 1993, the University of Delaware, in cooperation with the U.S. Geological Survey, initiated a pilot study to investigate the transport of atmospherically deposited trace elements through a pristine, forested watershed in the headwaters of the Potomac River (Bear Branch). This study, funded by the Maryland Department of Natural Resources' Power Plant Research Program, has the following specific objectives: (1) to accurately determine the wet and dry atmospheric trace element loads into the watershed, (2) to compare the total atmospheric load versus fluvial output of trace metals and (3) to estimate the transport of atmospherically deposited trace elements through the watershed relative to the trace metals naturally relapsed during weathering of the soil and rock within the study area. The Bear Branch watershed was chosen as it has been well-characterized hydrologically, is representative of the land use in the Potomac basin (60 percent of which is forested), and possesses an unreactive quartzite lithology which simplifies its geochemical weathering behavior. Further watershed transmission studies began in the spring of 1996 in the Appalachian Plateau of Western Maryland. While the results of these studies will represent an initial attempt to quantify the watershed retention/transport of atmospheric loads, further work is needed to extend the study to other regions with differing land use/geomorphology, in order to accurately determine an integrated, baywide watershed transport factor.

The next section describes programs in other coastal waters, as well as research relevant to atmospheric deposition in these coastal waters.

IV.D Coastal Waters

As stated previously, section 112(m) designates "coastal waters" as EPA's National Estuary Program (NEP) and NOAA's National Estuarine Research Reserve System (NERRS) estuaries. These two programs and EPA's Gulf of Mexico Program are the three significant coastal waters programs, outside the Chesapeake Bay Program, established in the last decade. Although the Gulf is not designated by name under section 112(m) of the CAA, 11 estuaries in various locations spanning the Gulf coastline are either NEP or NERRS (current or proposed) designated sites and, thus, are designated Great Waters.

The NEP, NERRS, and Gulf of Mexico programs differ in purpose and procedure, but they all serve to protect and restore the nation's valuable coastal water resources. The remainder of this section provides background information on each of these programs, followed by a discussion of studies of atmospheric deposition to coastal waters and future research needs.

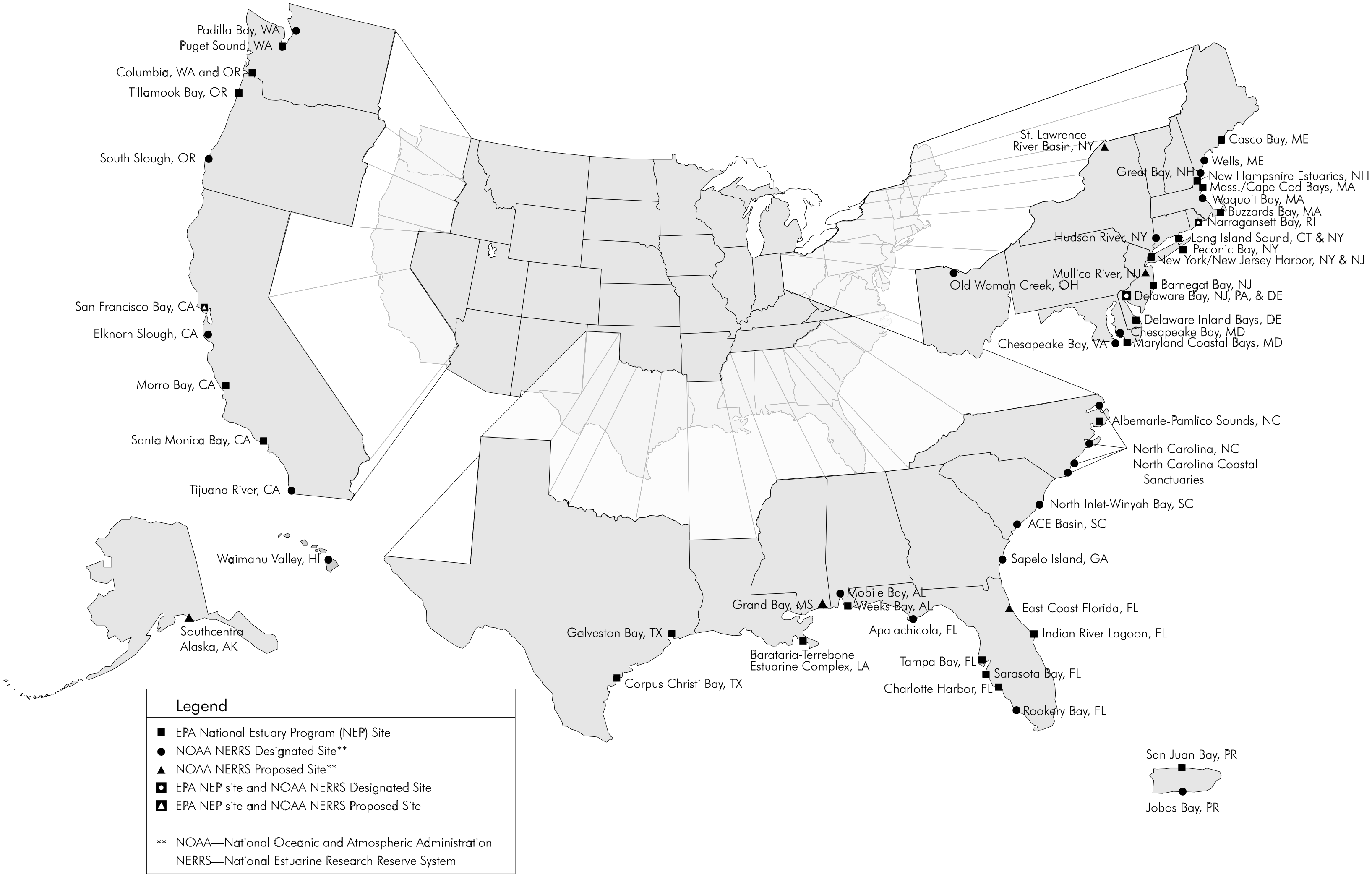
National Estuary Program

Congress established the National Estuary Program (NEP) in 1987 under section 320 of the Clean Water Act. Through the NEP, states nominate estuaries of national significance that are threatened or impaired by pollution, development, or overuse. EPA evaluates the nominations and selects those estuaries for which there is evidence of political support, citizen and government involvement (local, state, regional, federal), and available scientific and technical information to address the identified problems. For the selected estuaries, EPA convenes management conferences with representatives from all concerned groups (e.g., industry, agriculture, environmental organizations, state agencies) to more fully characterize problems and seek solutions through a collaborative decision-making process. Through these conferences, Comprehensive Conservation and Management Plans (CCMPs) are developed, for which EPA provides up to 75 percent of the funding. Each management conference must complete development of the CCMP within three to five years of the date the conference was convened. Upon approval of the CCMP, action plans are carried out by implementation agencies involved with development of the plan.

The purpose of the NEP is to identify nationally significant estuaries, protect and improve their water quality, and enhance their living resources. The NEP currently includes 28 estuaries (individually called NEPs) representing a wide spectrum of environmental conditions (see Figure IV-20). Because there are over 150 estuaries in the United States and only a small fraction can be targeted for action through the NEP, the NEP is intended to act as a national demonstration program, such that results and lessons learned in the NEP estuaries are shared and applied by parties concerned with other estuaries throughout the country. It should be emphasized that the NEP is a management program rather than a research program and relies on the research of other agencies and institutions to support its work. The development of support networks and cooperation between local, state, regional, and federal agencies is one of the program's greatest assets.

Several NEPs have identified atmospheric deposition of pollutants as a concern to the health of their estuaries. These NEPs have either initiated studies on the contribution of atmospheric deposition to annual loadings for nitrogen and/or other pollutants or expressed serious interest to EPA in conducting such projects. Nine NEPs submitted proposals to EPA in early 1996 for funding under section 112(m) of the CAA. To date, only two NEPs (Tampa

FIGURE IV-20
Locations of NEP and NERRS Sites



Bay and Galveston Bay) have received funding under section 112(m) to conduct such work. Other NEPs (Casco Bay, Delaware Bay, Long Island Sound, Massachusetts Bays, and Peconic Bay) have initiated exploratory studies funded by their own program and other sources. Atmospheric deposition research related to the NEPs is discussed later in this section.

National Estuarine Research Reserve System

Another program established to recognize the importance of estuaries is the National Estuarine Research Reserve System (NERRS), which was created by Congress in 1972 under the Coastal Zone Management Act and which operates under the authority of NOAA. The mission of NERRS is to establish and manage, through the cooperation of federal, state, and community efforts, a national system of estuarine research reserves that are representative of various regions and estuary types in the United States, in order to provide opportunities for long-term research, education, and stewardship.

The process for designating and maintaining a NERRS site includes five main activities, all of which may be partially funded by NOAA: (1) predesignation phase (includes selection of the site by the state and, after approval of the site by NOAA, preparation of a draft and final management plan and environmental impact study and completion of basic characterization studies); (2) acquisition of land and development activities; (3) after designation as an NERRS site, implementation of research, educational, and research programs detailed in the research reserve management plan; (4) estuarine research and monitoring; and (5) educational and interpretive activities.

Currently, 22 areas are designated as NERRS sites, including portions of Chesapeake Bay and associated lands in Maryland and Virginia (see Section IV.C for a detailed discussion of Chesapeake Bay). Six additional NERRS sites have been proposed or are in the beginning stages of development. See Figure IV-20 for the location of the NERRS estuaries.

Studies on the direct contribution of atmospheric deposition to NERRS waters are limited at this time. Available information on atmospheric deposition research related to NERRS waters is presented later in this section. For example, the indirect contribution of atmospherically deposited nitrogen to Waquoit Bay, Massachusetts, through its watershed has been estimated and modeled as part of the multi-year Waquoit Bay Ecological Risk Assessment Case Study (NOAA and MA DEM 1996; U.S. EPA 1996f).

Gulf of Mexico Program

The Gulf of Mexico is a very important resource to all of North America. Its surface area is about 1,603,000 km^2, large enough to cover one-fifth of the continental United States. The U.S. portion of the Gulf's shoreline measures over 2,500 km from the Florida Keys to the Rio Grande. Taking into account the shoreline length of all the bays, estuaries and other coastal features of the Gulf, its effective shoreline length is about ten times that amount. The 21 major estuaries along the Gulf coast account for 24 percent of all estuarine area in the 48 contiguous states, and 55 percent of the marshes. The watershed of the Gulf includes more than two-thirds of the continental United States (plus one-half of Mexico and parts of Canada, Guatemala, and Cuba), with the Mississippi River watershed alone draining about 40 percent of the continental United States.

The Gulf of Mexico Program (GMP) was established in 1988 in response to citizens' concerns over declines in the Gulf's fish, shellfish, and wildlife; the quality of life in many coastal communities; the need to protect the remaining valuable resources and prevent problems before they occur; and to forge a positive relationship between ecological health and economic vitality of the Gulf region. The GMP is a unique organization that involves representatives from government agencies (federal, state, and local), business and industry, non-profit organizations and educational institutions, and interested individuals in the process of setting environmental goals and implementing actions to achieve those goals. The aim is to foster coordination and cooperation among these organizations in order to reduce costs and coordinate actions.

The GMP is not a regulatory program, but rather an approach to environmental protection, similar to the Chesapeake Bay Program, that is founded on the principles of:

- Partnership among government agencies, private, and non-government interests to define and characterize concerns and implement solutions;
- Sound science and information as the basis of informed decision-making to guide actions; and
- Public involvement to determine goals, identify solutions, and generate the consensus needed for action.

Since its beginning, the GMP has made significant progress in effectively involving a broad spectrum of the public in defining goals and objectives and in characterizing fundamental issues. The fundamental goals of the GMP are to:

- Protect human health and the food supply;
- Maintain and improve Gulf habitats that support living resources (fish, shellfish, and wildlife); and
- Maintain and enhance the sustainability of the Gulf's living resources.

In the past few decades, the Gulf of Mexico has been degraded, largely due to nutrient enrichment and habitat loss. The contribution of atmospheric nitrogen to nutrient enrichment is not well understood and is possibly a significant concern. Fed by nutrient-enriched waters of the Mississippi River, a large area of near-bottom waters commonly become depleted in oxygen, or hypoxic. At its peak, this area (known as the "hypoxic zone") can extend over a 18,192 km^2 area from the coastal waters of the Mississippi River Delta of Louisiana to those of eastern Texas. Stresses to the benthic (bottom-dwelling) community have been observed in this zone, including mortality of larger non-swimming benthic organisms. This and other possible disruptions to the food chain threaten to affect the commercial and recreational fish species within the hypoxic area. In addition to the Louisiana Shelf hypoxic zone, 18 other coastal areas in the Gulf have experienced hypoxia due to increasing nutrient concentrations or loads. Evaluation of atmospheric deposition of pollutants to the Gulf is discussed below, including research in NEPs located in the Gulf, as well as two studies conducted in the Gulf as a whole.

Studies of Atmospheric Deposition in NEP and Other Coastal Waters

Nitrogen Loadings to Coastal Waters

At least 40 studies around the world, the majority of which have been published since 1990, have addressed at least the direct loading component of atmospheric nitrogen loadings. However, the measurement and modeling techniques used vary considerably among individual studies, making comparisons difficult. Table IV-11 presents a summary of selected studies performed along the East and Gulf coasts of the United States that are comparable in broad terms. The two criteria for selecting these studies were that the study results were either published in a credible peer-reviewed journal or advocated by a major management organization (e.g., an EPA NEP). These studies can be divided into two groups: those that considered both direct and indirect nitrogen loads and those that considered only direct loads. Data from these studies show that, in general, the amount of atmospheric nitrogen input is related to the size of the waterbody and its watershed. To some extent, the percent load from atmospheric deposition is influenced by whether both direct and indirect deposition were considered.

Municipal and industrial wastewater discharges and urban runoff/storm sewer inputs have historically been considered the largest sources of pollutants to coastal waters. Recently, however, researchers have begun to investigate the role of atmospheric deposition as a source of pollutants in a coastal waters (e.g., Paerl 1985, 1993; Scudlark and Church 1996). Assessing the impact of atmospheric deposition of pollutants has become a priority for many NEPs and other coastal watershed protection programs. There is a clear need to characterize the types, quantities, and sources of pollutants that are being directly and indirectly deposited from the atmosphere into these estuaries. Recent studies on atmospheric deposition to coastal waters are discussed briefly below and are presented in Tables IV-12 and IV-13. Data as of December 1995 on the contribution of atmospheric deposition to nitrogen loadings to Chesapeake Bay and other coastal waters are presented in Table IV-11; in this table, information is presented first for Chesapeake Bay and a few related tributaries, followed by other coastal waters in descending order of tidal water area. Data on the contribution of atmospheric deposition to the loadings of toxic pollutants in coastal waters are presented in Table IV-12; the coastal waters are listed in geographical order clockwise from the northeast coast to the northwest coast. In general, the studies discussed below have evaluated the relative contribution of nitrogen and other pollutants of concern, and do not attempt to identify the particular emission sources contributing to this pollution.

As mentioned above, research on atmospheric deposition to Tampa Bay, Florida, and Galveston Bay, Texas, has been conducted under the Great Waters program. The Tampa Bay and Galveston Bay studies are discussed first, followed by a description of other studies of atmospheric deposition to NEP estuaries (i.e., those that have been funded through sources other than EPA's Great Waters program). Initial observations from these studies suggest that direct and indirect loadings from air deposition may be significant sources of nitrogen and toxic pollutants to coastal waters.

Tampa Bay. As recently as 1991, atmospheric deposition of nitrogen, air toxics, and other pollutants was assumed to have a minimal effect on water quality in Tampa Bay. However, based on a methodology developed by the Environmental Defense Fund (Fisher et al. 1988), early calculations provided an early indication of likely nitrogen loadings from atmospheric deposition in Tampa and Galveston Bays.

TABLE IV-11
Estimates of Atmospheric Nitrogen Loadings to Selected Coastal Waters[a]
(in millions of kg)

Coastal Water	Surface Area (km^2)		Deposition to Watershed	Direct Deposition to Tidal Waters	Indirect Atmos. Load From Watershed	Total Atmospheric Load	Total Load From All Sources	% Load from Atmosphere	Reference[b]
	Watershed	Tidal Waters							
Chesapeake Bay (MD/VA)	165,886	11,400	175	16	29	45	170	27	5
Rhode River (MD)	33	4.9	—	0.005	—	0.005	0.012	40	6
Choptank River (MD)	1,779	361	—	0.57	—	0.57	1.54	37	11
Patuxent River (MD)	2,393	137	—	0.22	—	0.22	12.6	13	11
Potomac River (MD)	29,940	1,210	—	1.9	—	1.9	35.5	5	11
New York Bight (NY/NJ)[c]	50,107	38,900	69	54	8	62	164	38	1
Albemarle-Pamlico Sound (NC)	59,197	7,754	~39	3.3	6.7	10	23	44	4
Long Island Sound (NY/CT)	43,481	4,820	43	5	6	11	60	20	3
Massachusetts Bays (MA)	—	3,700	—	1.6-6	—	1.6-6	22-30	5-27	15
Delaware Bay (DE)	36,905	1,846	53	3	5	8	54	15	2
Tampa Bay (FL)	6,216	1,031	—	1.1	—	1.1	3.8	28	14
Guadalupe Estuary (TX)	—	551	—	0.31	—	0.31	4.2-15.9	2-8	13
Narragansett Bay (RI)	4,708	328	4.2	0.3	0.3	0.6	5	12	1
			—	0.4	—	0.4	9	4	12
Newport River Coastal Waters (NC)	340	225-1,600	—	0.095-0.68	—	0.095-0.68	0.27-0.85	36-80	4
Sarasota Bay (FL)	524	135	—	0.16	—	0.16	0.6	26	10
Delaware Inland Bays (DE)	800	83	—	0.28	—	0.28	1.3	21	9
Flanders Bay (NY)	83	39	—	0.027	—	0.027	0.36	7	8
Waquoit Bay (MA)	~70	~8	0.062	—	0.0065	0.0065	0.022	29	7

[a] Estimates as of December 1995.

[b] (1) Hinga et al. 1991; (2) Scudlark and Church 1993; (3) Long Island Sound Study; (4) Paerl and Fogel 1994; (5) Linker et al. 1993; (6) Correll and Ford 1982; (7) Valiela et al. 1996; (8) Peconic Bay NEP; (9) Delaware Bays NEP; (10) Sarasota Bay NEP 1995; (11) Boynton et al. 1995; (12) Nixon et al. 1995; (13) Brock et al 1995; (14) Tampa Bay NEP, Zarbock et al. 1994; (15) Massachusetts Bays NEP 1996.

[c] New York Bight extends from Cape May, New Jersey, to Long Island Sound.

Source: Adapted from Valigura et al. 1996.

TABLE IV-12
Studies of Atmospheric Loadings of Toxic Pollutants to NEP Coastal Waters

Coastal Water	Pollutants of Concern Evaluated[a]	Relative Contribution of Atmospheric Deposition for the Great Waters Pollutants of Concern	Reference
Massachusetts Bays (MA)	PAHs, PCBs, cadmium, lead, mercury	Direct atmospheric deposition estimated to contribute: PAHs, 9-46%; PCBs, 28-82%; cadmium, 17-31%; lead, 39-45%; mercury, 4-13%.	Menzie-Cura & Associates 1991
Narragansett Bay (RI)[b]	PCBs, PAHs	Direct atmospheric deposition found to contribute 3% of PCBs and 12% of PAHs.	Latimer 1997
New York-New Jersey Harbor Estuary and Bight (NY/NJ)	Cadmium, lead, mercury, PCBs, dioxins, PAHs, various pesticides	Atmospheric deposition identified as a significant contributor to total pollutant loading for lead (39-54%), but may have been over-estimated. For other pollutants, either atmospheric deposition was insignificant or estimates were not developed.	NY-NJ Harbor NEP 1995
Delaware Bay (DE/NJ/PA)[b]	Lead, mercury, PCBs, various pesticides, volatile organic compounds (VOCs)	Atmospheric deposition (both direct and indirect) found to be a significant source of mercury (80%) and PCBs (34%). For lead, atmospheric deposition contributed less than 5%. For other pollutants, either atmospheric deposition was insignificant or estimates were not developed.	Frithsen et al. 1995b
Tampa Bay (FL)	Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs	Direct and indirect atmospheric deposition identified as a significant contributor of cadmium (46%), lead (12%), and PCBs[c], but not a significant source of chlordane, DDT, dieldrin, or mercury (1% each). Estimates for PAHs were not developed.	Frithsen et al. 1995a
	Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs	On-going study - no results yet.	U.S. EPA 1995f
Galveston Bay (TX)	PAHs, PCBs, selected pesticides, lead, cadmium, mercury	On-going study - no results yet.	U.S. EPA 1995g
Santa Monica Bay (CA)	PAHs, metals, chlorinated organics	Atmospheric deposition was estimated to be a significant source of lead and PAHs.	SMBRP 1994

[a] For a discussion of other pollutants evaluated, study methods, and uncertainties, see referenced study.
[b] These NEPs also are NERRS designated sites.
[c] Estimates of PCB loadings could be made for atmospheric deposition only; therefore, a relative comparison to other sources could not be made.

A recent study of nutrient (i.e., nitrogen and phosphorus) and suspended solids loadings conducted for the Tampa Bay NEP suggests that direct atmospheric deposition of nitrogen to the tidal waters of Tampa Bay is the second largest source of nitrogen entering the Bay, contributing up to 28 percent of the total nitrogen load (Zarbock et al. 1994). The largest source of nitrogen, according to that study, is urban storm water runoff. A portion of the nitrogen entering the Bay from urban storm runoff represents atmospherically deposited nitrogen to impervious surfaces such as paved roads and sidewalks. These studies prompted the Tampa Bay NEP to revise its CCMP to consider atmospheric deposition issues.

Another study, conducted by the Tampa Bay NEP (Frithsen et al. 1995a), investigated the contribution of atmospheric deposition relative to point sources, urban runoff, and other nonpoint sources for specific chemical contaminants of concern. The contaminants, which were selected based on their potential for toxic effects and their concentrations observed in sediment samples, included: six metals (cadmium, chromium, copper, lead, mercury, and zinc); four organochlorine pesticides (chlordane, DDT, dieldrin, and endrin); and two classes of organic chemicals (polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs)). In addition, loadings were estimated for arsenic and iron because of the potential of these chemicals have for environmental harm or interaction with contaminants of concern through chemical processes. Because the study used numerous information sources, representing a wide range of spatial and temporal conditions, a great deal of uncertainty exists regarding the absolute estimates of atmospherically deposited chemical contaminants to Tampa Bay. Study results are intended to establish the relative magnitudes of different classes of sources in order to set priorities for more detailed research and monitoring activities. Some general conclusions, however, are apparent:

- Contaminant inputs to Tampa Bay from runoff and transfer of atmospherically deposited contaminants to the watershed (i.e., indirect loading) are approximately two-thirds the contribution of contaminant inputs from direct deposition to tidal waters (i.e., direct loading);

- Atmospheric deposition contributes a sizeable percentage to total annual load for the following metals: cadmium (46%), copper (18%), chromium (13%), lead (12%), and iron (11%);

- Atmospheric deposition of mercury is around 4 kg/yr and atmospheric deposition of pesticides is estimated as 10 kg/yr, each representing about one percent of the total load; and

- Atmospheric deposition is the only pathway that contributed a measurable amount of PCBs, estimated as a minimum total load of 11 kg/yr. No estimate could be developed for PAHs using available datasets.

Ongoing monitoring, described below, conducted by the Tampa Bay NEP, local governments, and other collaborators will better define the spatial distribution of atmospherically deposited chemical contaminants and nitrogen throughout the 10 major basins of Tampa Bay and its watershed as well as the relative contributions of local, regional, and global emission sources.

An ongoing cooperative study administered by the Tampa Bay NEP will substantially expand air transport and deposition monitoring and modeling projects in an effort to develop nationally recognized quantitative assessments for important air deposition parameters. Now in its second year of operation, the Tampa Bay Atmospheric Deposition Study (TBADS) involves EPA's Great Waters program; the local governments in Hillsborough, Pinellas, and Manatee Counties; the Florida Department of Transportation; the Southwest Florida Water Management District; Florida State University; a private consultant; and other organizations. TBADS will attempt to determine: (1) what fraction of the nitrogen and toxic pollutants emitted annually by specific sources within the Tampa Bay watershed enter the Bay waters (i.e., are deposited either onto the water surface directly or onto the watershed and subsequently enter the Bay waters through runoff); and (2) what are the relative contributions of local sources (i.e, inside the watershed) versus remote sources (i.e, outside the watershed) to atmospherically deposited nitrogen and toxic pollutants in the watershed. Projects that have been initiated to address elements of this air deposition program include:

- Intensive daily wet and dry deposition monitoring at Gandy Site, located on Tampa Bay's Interpeninsula, for nitrogen and toxic pollutants for an additional 12 months, yielding two years of continuous data collected and analyzed according to AIRMoN protocol;

- Application (with the Florida State University Center for Tropical Meteorology) of a regional air mass movement model developed by Pennsylvania State University (Penn State/NCAR Mesoscale Model Version 5) to investigate air transport at a coarse grid for the southeastern United States while maintaining a much higher resolution grid over the Tampa Bay area to estimate retention times for air masses within the watershed under different meteorological conditions;

- Integration of NOAA's Physical Oceanographic Real-Time System (PORTS) Meteorological data collected at several over-water stations (with NOAA and the University of Florida); and

- Intensive stormwater sampling to measure stormwater runoff, nitrogen, and toxic contaminant concentrations and loads at up to four gaged subbasins. Data will be used to estimate the relative contribution of atmospheric loading to stormwater for different land use types (residential, urban, industrial, and/or urban parks); it is expected that this transfer coefficient information will be useful not only to Tampa Bay and Florida, but also to watersheds nationwide.

An important element of the Tampa Bay atmospheric deposition program is the participation and coordination of local and federal government programs and state agencies with the Great Waters program and the Tampa Bay NEP.

Galveston Bay. The Great Waters program conducted a screening atmospheric deposition monitoring program in Galveston Bay, Texas, which was chosen as the site to establish monitoring for the Texas Regional Integrated Atmospheric Deposition Study (TRIADS) as a representative of a Gulf of Mexico estuary. Monitoring at the TRIADS site began in February 1995. To facilitate comparability, the sampling and analytical design of TRIADS is similar to that of existing monitoring sites in the Great Lakes and Chesapeake Bay. The goals of this study are to evaluate the contribution of atmospheric deposition of selected contaminants to the Bay and to evaluate long-range transboundary transport of contaminants. Pollutants measured include cadmium, lead, mercury, nitrogen, PAHs, PCBs, and selected pesticides. Results from TRIADS complement and add to data from other investigations in Galveston Bay, including studies by the Galveston Bay NEP, EPA's Environmental Monitoring and Assessment (EMAP) program, NOAA National Status and Trends (NS&T) program, and special urban-pollutant studies in Houston, Texas. Data from these programs and the TRIADS data will be used to estimate the cumulative, direct and indirect impacts of atmospheric deposition to pollution of Galveston Bay.

Although early calculations suggested that atmospheric deposition could be a significant contributor to nitrogen loads delivered annually to Galveston Bay, the relevance of this finding to the health of the ecosystem for Galveston Bay is not as obvious as for either Chesapeake Bay or Tampa Bay. While all three estuaries have experienced declines in submerged aquatic vegetation (SAV), studies on SAV in Galveston Bay are limited in contrast to documented cases of large-scale changes in other major estuaries (Pulich et al. 1991). While atmospheric nitrogen loading may contribute to the incidence of hypoxia, other factors appear to be causing this phenomenon, which is quite localized in Galveston Bay compared to its manifestation in the other two estuaries.

In contrast to the perceived limited biological effects from atmospheric deposition of nitrogen, previous research has suggested that atmospheric deposition of toxic contaminants may be affecting fish and shellfish in Galveston Bay, and thus contributing to human health risk. A pilot study performed for the Galveston Bay NEP documented the presence of dioxins, furans, lead, mercury, PAHs, PCBs, and pesticides in certain species of finfish and shellfish, but could not determine the sources of these contaminants (Brooks et al. 1992). Monitoring data from the TRIADS site detected the presence of all these chemical contaminants in air samples, suggesting that atmospheric deposition may be a significant source (Battelle 1995). Continued monitoring will enable scientists and managers to more fully evaluate this problem and determine the relative effect of atmospheric deposition versus point and nonpoint source inputs into Galveston Bay.

Casco Bay. The primary pollutants of concern for atmospheric deposition to Casco Bay, Maine, include PAHs, PCBs, nitrogen, phosphorus, sulfates, pesticides, and mercury and other trace metals. Recent sediment studies have found elevated concentrations of some pollutants (i.e., cadmium, lead, mercury, PAHs, PCBs, silver, and zinc) near population centers and waste discharges, but also observed elevated levels in rural eastern Casco Bay away from these known sources (Wade et al. 1995). A circulation model study of the Bay did not clearly indicate any possible sources for these pollutants, suggesting atmospheric deposition as a significant source (Pearce et al. 1994). While elevated levels of lead found in Casco Bay sediments were relatively near potential sources, elevated levels of cadmium were found far from any known local source. A deposition study would provide empirical verification of processes believed to be occurring at Casco Bay.

Delaware Bay. In Delaware Bay, studies have shown that direct and indirect atmospheric deposition provide 15 percent of the annual nitrogen input, increasing to 25 percent in late spring and early summer (Scudlark and Church 1993). The relative nitrogen loading is slightly lower than observed in nearby Chesapeake Bay (27 percent), and much lower than in the Delaware Inland Bays (Rehoboth and Indian River Bays) where direct atmospheric deposition alone contributes 27 percent of the total nitrogen load (Cerco et al. 1994). The contribution to the Delaware Bay is lower because of higher point source nitrogen loading to Delaware Bay and the influence of a highly urbanized watershed.

As part of a Delaware Estuary Program study to estimate contaminant inputs, atmospheric deposition was found to be a significant source of mercury (80 percent) and PCBs (34 percent) (Frithsen et al. 1995b). As is the case in other regions, more research is warranted on atmospheric inputs of mercury and the resulting effects on estuarine and human health. To evaluate the effect atmospheric deposition of mercury has on the Delaware, Rehoboth, and Indian River estuaries, a precipitation monitoring station was established at Lewes, Delaware, in 1995 in conjunction with EPA's National Atmospheric Deposition Program Mercury Deposition Network.

Gulf of Mexico. It is probable that significant amounts of nitrogen are deposited into the Mississippi River Basin via atmospheric deposition, but there has been little investigation conducted regarding atmospheric nitrogen as a source of nitrogen for the Mississippi River drainage basin. Some basic estimates using National Atmospheric Deposition Program (NADP) data were provided at a Hypoxia Conference held by the Gulf of Mexico Program in August 1996. These estimates showed significant variability in quantity of atmospherically deposited nitrogen, with a range of 0.55 million to 3.08 million tons. This variability is due to differing assumptions of what atmospheric nitrogen input sources should be included, what forms of deposition should be measured (e.g., dry deposition), and what nitrogen compounds should be analyzed (e.g., ammonium). At the high end, atmospheric nitrogen would be on par with animal manure, ranking as the second highest source of nitrogen input to the watershed. At the lower end, estimated atmospheric nitrogen inputs would rank as the fifth highest source of nitrogen input for the watershed. This wide variability in estimated quantity points to the need for further and more refined estimates of atmospheric nitrogen inputs to the Gulf.

Hypoxia Conference

A conference was convened in 1996 in response to the hypoxia problem in the Gulf of Mexico. Topics addressed included:

- Characterization of the hypoxic zone;
- Economic impacts and trends in fisheries attributable to the hypoxia;
- Causes of the hypoxic zone;
- Sources and delivery of nutrients in the watershed, including atmospheric deposition;
- Current efforts to control nutrient loads; and
- Information and policy required for action.

Long Island Sound. A chronic problem in Long Island Sound is the low oxygen levels (hypoxia) that are observed during the summer. An early study noted that excess nitrogen loading was a major cause of hypoxia and estimated that atmospheric loading directly to the water surface contributed 8 percent of the total nitrogen delivered to the Sound (LIS Study 1990). A later study produced essentially the same estimate for the total contribution of direct atmospheric deposition, but divided it into two components: an amount from "natural" sources (i.e., background approximating "amount believed to have been delivered to Long Island Sound in pre-Colonial days") and an amount from "human-induced" sources (LIS Study 1994). Using measurements of wet and dry deposition from two sites along the Connecticut shore, Miller et al. (1993) estimated direct atmospheric loadings to Long Island Sound. The Long Island Sound Study used these data and literature values to develop the estimates shown in Table IV-12, and concluded that (direct and indirect) atmospheric deposition may be responsible for 17 to 24 percent of total nitrogen entering the Sound.

In the most recent study, all sources of nitrogen, including atmospheric deposition, are divided into "natural" and "human-caused" components (Stacey 1996). This study concluded that atmospheric deposition from human activities in the New York and Connecticut portions of the Long Island Sound watershed accounts for 13.6 percent of the total enriched or "human-caused" load. Further work is necessary to model relationships among air quality, direct and indirect atmospheric deposition, and runoff concentrations to receiving waters of the Sound.

Massachusetts Bays. In one Massachusetts Bays NEP study, direct atmospheric deposition was estimated to contribute 5 to 16 percent of total nitrogen load to Massachusetts Bays (Menzie-Cura & Associates 1991). In another Massachusetts Bays NEP study, direct atmospheric deposition of nitrogen was estimated to account for 6 to 8 percent of total nitrogen loadings to the Bays (Zemba 1995). Different methodologies were used to estimate nitrogen loadings in these two studies. The estimate by Zemba (1995) used literature values and ten years (1981-1991) of wet deposition data from the NADP. Other studies cited by the Massachusetts Bays NEP suggest that the contribution of atmospheric deposition may be higher, about 16 to 20 percent of total nitrogen load, excluding exchange with the Gulf of Maine (Massachusetts Bays NEP 1996).

Atmospheric deposition is also a significant contributor of organic pollutants and trace metals to Massachusetts Bays. Menzie-Cura (1991) estimated that direct atmospheric deposition was a significant source of PAHs (9-46 percent), PCBs (28-82 percent), cadmium (17-31 percent), and lead (39-45 percent). A subsequent Massachusetts Bays NEP study generally corroborated the Menzie-Cura (1991) metal deposition results, although lead deposition rates were slightly lower (Golomb et al. 1995). The lead deposition estimates may be lower in Golomb (1995) because the data used in the Menzie-Cura study were obtained prior to the phase out of leaded gasoline. Golomb (1995) also indicated that PAH deposition may have been underestimated and that PCB concentrations were below detection limits and, therefore, atmospheric deposition rates for PCBs were not calculated. Because PCB concentrations were below the detection limit, more precise field measurements of wet and dry deposition of PCBs are necessary to verify the initial estimates and to determine the relative impact of atmospheric deposition of PCBs to Massachusetts Bays.

Peconic Bay. Nitrogen from atmospheric deposition to the Peconic River and Flanders Bay, New York, is estimated to be about 73 kg/day, or about five percent of the total nitrogen loading to that area (Suffolk County 1992). The impact of atmospheric deposition on eutrophication in Peconic River and Flanders Bay is considered to be relatively small in relation to other point and nonpoint sources. Atmospheric deposition is believed to be much more significant in terms of relative eutrophication impacts to Peconic Estuary surface waters east of Flanders Bay. Detailed loading estimates for these eastern areas, as well as for specific subwatersheds, are being developed for the Peconic NEP waters, and the relative eutrophication impacts of sources are being assessed through computer modeling.

Future Research Needs in NEP and Other Coastal Waters

Research on atmospheric deposition to coastal waters has been limited to a few areas, and most studies have focused on identifying and determining the concentration of pollutants of concern in water and sediment, and measuring concentrations of pollutants in precipitation. Due to limited funding, many preliminary NEP studies are restricted to using historical data to estimate atmospheric deposition. Some NEP studies have used a rough mass balance approach to determine the relative loading of each pollutant to the estuary, but more precise quantitative mass balances are needed, which require accurate and comprehensive atmospheric data.

Establishing the total contribution of pollutants and their sources is an important part of developing and implementing CCMPs for NEPs, and the lack of knowledge about the concentrations, deposition, and potential sources of airborne pollutants makes sound policy formation for the estuaries difficult. The question of the magnitude of pollutant deposition from the air has become more important as other sources of pollution to rivers, lakes, streams, and coastal waters have been identified and significantly reduced.

Research questions for the NEP estuaries and other coastal waters include:

- What are the concentrations and loadings of pollutants that are being supplied by atmospheric deposition?

- What are the relative contributions of these inputs to the total load of pollutants entering the estuary?

- What are the emission sources that affect the estuaries and where are they geographically located?

- Does atmospheric deposition (direct and indirect) of contaminants cause or contribute to biological harm in benthic (bottom-dwelling) or pelagic (suspended, planktonic, or water column) communities, or affect human health?

- What economically and technically feasible methods are available to effectively reduce airborne pollutants and their effects on estuaries?

The NEP estuaries provide an excellent opportunity to evaluate the effects and contribution of atmospheric deposition of contaminants to a varied set of ecological, environmental, and anthropogenic conditions. The NEP also provides a "grassroots" forum for addressing and correcting regional and national air quality issues as they pertain to our coastal waters. Recommendations for further atmospheric deposition research in coastal waters to help answer the above questions include:

- Utilize existing databases and ongoing work of established research programs and coordinate research initiatives with these programs;

- Protect and enhance existing monitoring programs;

- Establish long-term water and air quality monitoring programs that incorporate sampling for atmospheric deposition of contaminants for a subset of NEP estuaries representing various geographical regions and environmental conditions;

- Use sampling data from monitoring programs to track trends and spatial variability to develop more accurate loading estimates;

- Coordinate efforts between NEP estuaries and other Great Waters program studies to identify local, regional, and national sources of airborne pollutants;

- Pursue detailed atmospheric chemistry and deposition models for estimating atmospheric deposition to NEP estuaries;

- Develop a multi-party effort to identify and demonstrate appropriate pollution prevention techniques;

- Apply existing atmospheric circulation models to fill in data gaps between measured and estimated atmospheric deposition and to aid in tracing the pollutants in the estuaries back to their probable sources; and

- Support process-related research to establish cause and effect relationships between atmospheric deposition of contaminants and alterations of water quality, fisheries, recreational and other economic and ecological resources of receiving estuarine and coastal waters.

This research is needed not only to assist decision-makers for specific coastal waters, but to form a comprehensive picture of atmospheric deposition across the United States. In addition, coordinated use of other mechanisms, such as voluntary pollution prevention, can help control the negative impacts of atmospheric deposition to water quality in NEP estuaries, especially at the local and regional level.